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Air Toxics Emissions from Motor Vehicles

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Technical Report

Air Toxics Emissions From Motor Vehicles

By

Penny M. Carey

September, 1987

NOTICE

Technical Reports do not necessarily represent final EPA decisions or positions. They are intended to present technical analysis of issues using data which are currently available. The purpose in the release of such reports is to facilitate the exchange of technical information and to inform the public of technical developments which may form the basis for a final EPA decision, position or regulatory action.

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Preface

This report titled "Air Toxics Emissions from Motor Vehicles" is designed to be a compilation of available information on emission levels of potentially carcinogenic substances from motor vehicles. While earlier EPA reports (e.g., "The Air Toxics Problem in the United States; An Analysis of Cancer Risks for Selected Pollutants") discussed air toxics emissions in general, their main emphasis was not mobile sources such as in this report.

EPA currently plans no further regulatory action as a result of this report. EPA invites comments on this report and plans to update this information as additional data become available.

A draft version of this report was circulated prior to the release of this final version. Users are cautioned that Section 7.0 dealing with gasoline PIC/POM/organics has been revised to correct an error due to a misinterpretation of some of the references. The estimated risks from gasoline organics are now much lower although still in the range of 100-200 cancers/year. The summary tables have been changed accordingly. This is the only substantive change of note from the draft version.

TABLE OF CONTENTS

Page

PREFACE i

EXECUTIVE SUMMARY v

1.0 INTRODUCTION 1

2.0 DIESEL PARTICULATE 2
  2.1 Formation and Composition 2
  2.2 Control Technology 3
  2.3 Emissions 4
    2.3.1 Emission Standards 4
    2.3.2 Emission Factors by Model Year 6
    2.3.3 Emission Factors for Calendar Years 1986 and 1995 6
    2.3.4 Nationwide Diesel Particulate Emissions. . 12
    2.3.5 Other Sources of Particulate 17
  2.4. Ambient Concentrations of Diesel Particulate . . 17
    2.4.1 Estimation of Urban and Rural Exposure . . 17
    2.4.2 Comparison to Other Exposure Estimates . . 24
      2.4.2.1 1983 OMS Exposure Estimate 24
      2.4.2.2 1981 Lovelace ITRI Exposure Estimate 25
      2.4.2.3 Exposure Estimate Based on a Lead Surrogate Approach 26
    2.4.3 Comparison to Particulate Monitoring Data. 27
  2.5 Health Effects of Diesel Particulate and Unit Risk Estimates 27
  2.6 Current and Projected Health Risk. 28

3.0 FORMALDEHYDE 30
  3.1 Formation, Composition and Control Technology. . 30
  3.2 Emissions 30
    3.2.1 Emission Factors for Calendar Years 1986 and 1995 30
    3.2.2 Nationwide Mobile Source Formaldehyde Emissions 33
    3.2.3 Other Sources of Formaldehyde 33
  3.3 Ambient Concentrations of Formaldehyde Emitted by Mobile Sources 34
    3.3.1 Estimation of Urban and Rural Exposure . . 34
    3.3.2 Contribution of Mobile Sources to Ambient Formaldehyde Levels 36
  3.4 Health Effects of Formaldehyde and Unit Risk Estimates 38
  3.5 Current and Projected Health Risk 39
  3.6 Current Activities 41

4.0 BENZENE 43
  4.1 Formation, Composition and Control Technology . 43
  4.2 Emissions 43
    4.2.1 Emission Factors for Calendar Years 1986 and 1995 43

TABLE OF CONTENTS (Continued)

Page

4.2.2 Contribution of Mobile Sources to Nationwide Benzene Emissions 45
4.3 Health Effects of Benzene and the Unit Risk Estimate 45
4.4 Current and Projected Health Risk 46
4.5 CARB Analysis of Current and Projected Health Risk 48
4.6 Comparison of EPA and CARB Health Risks 49
4.7 Current Activities 50

5.0 GASOLINE VAPORS 51

6.0 GAS PHASE ORGANICS 54
6.1 Formation and Control Technology 54
6.2 Composition 54
6.3 Mutagenicity of VOC 56
6.4 Risk Associated with Individual VOC 56
6.5 Reactivity of VOC 63
6.6 Current Activities 65

7.0 ORGANICS ASSOCIATED WITH NON-DIESEL PARTICULATE . . . 66
7.1 Emission Rates and Composition 66
7.2 Risk from Particle-Associated Organics 66

8.0 DIOXINS 73
8.1 Composition 73
8.2 Emissions 73
8.3 Concentrations of Dioxins 73
8.4 Current Activities 73

9.0 ASBESTOS 74
9.1 Emissions and Ambient Concentrations 74
9.2 Cancer Risk 74
9.3 Current Activities 74

10.0 VEHICLE INTERIOR EMISSIONS 76
10.1 Composition and Concentration 76
10.2 Cancer Risk and Current Activities 76

11.0 METALS 77
11.1 Lead 77
11.1.1 Source and Emission Factors 77
11.1.2 Health Effects 78
11.2 Manganese 80
11.2.1 Source and Emission Factors 80
11.2.2 Health Effects 81

TABLE OF CONTENTS (Continued)

Page

11.3 Platinum 82
11.3.1 Source and Emission Factors 82
11.3.2 Health Effects 82
11.4 Cadmium 83
11.4.1 Source and Emission Factors 83
11.4.2 Health Effects and Risk Estimate 83

12.0 SIX MONTH STUDY: SUMMARY AND COMPARISON OF RESULTS . 84
12.1 Purpose of Six Month Study and Summary of Results 84
12.2 Comparison of Six Month Study Results with Results of This Study 86
12.2.1 Formaldehyde 87
12.2.2 Benzene 89
12.2.3 PIC, B(a)P, Diesel Particulate and Gasoline PIC/POM 90
12.2.4 Gasoline Vapors 91
12.2.5 1,3-Butadiene 92
12.2.6 Ethylene 92
12.2.7 Asbestos 93
12.2.8 Ethylene Dibromide (EDB) and Cadmium . . . 93
12.2.9 Total Aggregate Risk 94

13.0 SUMMARY AND LIMITATIONS 95

REFERENCES 97

GLOSSARY OF TERMS 105

EXECUTIVE SUMMARY

EPA completed a report in 1985, commonly referred to as the Six Month Study, which contains estimated cancer risks for a number of toxic air pollutants. The report indicated that mobile sources may be responsible for a large portion of the aggregate cancer incidence. The Six Month Study, however, was broad in nature with the goal to obtain a quick assessment of the air toxics problem in the United States and to guide further studies.

The purpose of this report is to focus on air toxics emissions from mobile sources. Specific pollutants and pollutant categories which are discussed include diesel particulate, formaldehyde, benzene, gasoline vapors, gas phase organics, organics associated with non-diesel particulate, dioxins, asbestos, vehicle interior emissions and metals. The report considers all air carcinogens for which EPA has unit risk estimates and are emitted from motor vehicles. Where adequate information was available, quantitative estimates of cancer incidence were made for calendar years 1986 and 1995. The results were then compared to the results obtained in the Six Month Study.

The unit risks used in this paper are defined as the individual life time excess cancer risk from continuous exposure to 1 ug carcinogen per m^3 inhaled air. Assuming a life time is 70 years, the excess lung cancer risk in 1 year is derived by simply dividing the unit risk by 70. Using this approach, latency is ignored. The unit risks used in this paper are 95 percent upper confidence limits rather than best estimates. This is consistent with current EPA practice. The risk estimates presented should therefore be considered upper bound estimates.

The risks obtained in this study are summarized in Table S-1. The aggregate risk in 1986 for the total U.S. population is estimated to range from 385 to 2286 cancer incidences and drops roughly 40 percent by 1995. Reasons for the projected decrease in risk in 1995 include 1) more stringent diesel particulate standards for both light- and heavy-duty vehicles and 2) the increasing use of 3-way catalyst-equipped vehicles coupled with the phase out of non-catalyst-equipped vehicles.

As seen in Table S-1, there is a wide range of risk estimates associated with each pollutant. For diesel particulate, the range is due to the range of potency (or unit risk) estimates which were used and, for 1995, a range of assumptions regarding future diesel sales. The range for formaldehyde can be attributed to the uncertainty regarding the

Table S-1

Summary of Risk Estimates Contained in This Study[a]

Motor Vehicle Pollutant	U.S. Cancer Incidences/Year[b] 1986	1995[c]	Section of Report Discussing Pollutant
Diesel Particulate	178- 860	92-576	2
Formaldehyde	46- 131	29- 77	3
Benzene	92- 223	57-145	4
Gasoline Vapors	65	ND[d]	5
Other Gas Phase Organics			
1,3-Butadiene	0- 656	0-460	6
Ethylene	0- 60	0-31	6
Gasoline PIC/POM	1.3 - 176	0.78-136	7
Dioxins	ND	ND	8
Asbestos	0.41- 113.4	ND	9
Vehicle Interior Emissions	ND	ND	10
Cadmium	0.18	0	11
Ethylene Dibromide	1.8	0.54	12
Total:	385-2286	179-1426	

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[a] The risk estimates are 95% upper confidence limits.

[b] The risk estimates for gasoline vapors, asbestos, cadmium and ethylene dibromide are for urban exposure only. Risks for the other pollutants include both urban and rural exposure.

[c] The total risk in 1995 is slightly underestimated. Due to inadequate information and the sensitivity of 1995 risk to control decisions which have not yet been made, projected risk estimates were not made for some of the pollutants.

[d] ND=Not Determined.

contribution of photochemically formed formaldehyde. The low end of the range attempts to account only for formaldehyde directly emitted from the exhaust of motor vehicles. The high end of the range attempts to account for both formaldehyde directly emitted and formaldehyde formed in the atmosphere from other mobile source volatile organic compound (VOC) emissions. Both ends use a single unit risk estimate which is based on formation of malignant tumors.

For benzene, the lower limit is based on ambient concentrations predicted by a model, whereas the upper limit is based on actual monitoring data, with a mobile source fraction assigned based on the mobile source emissions contribution. The ranges for 1,3-butadiene and asbestos account for uncertainties in emission factors; for ethylene, the uncertainty is based on the unit risk estimate. The range for gasoline products of incomplete combustion (PIC) is due to a number of different assumptions regarding both emission factors and unit risk estimates.

Mobile source emissions are extremely complex. Hundreds of compounds, both in the gas phase and associated with particles are present. The lack of emissions data and/or health data and/or exposure data prevented quantitative risk estimates for any additional pollutants. Of particular concern are pollutants which are formed photochemically from mobile source emissions. This category of pollutants could have considerable impact but not enough is known to make a quantitative estimate.

A comparison of the results obtained in this study with those obtained in the Six Month Study is given in Table S-2. In the Six Month Study, the mobile source contribution to the air toxics risk was determined based on dispersion modeling of emissions in 35 highly populated counties (total population of 45 million). They represent a variety of industrial and population distributions, but are not considered a statistically representative sample of the country. When comparing the results of the Six Month Study with the results of this study, cancer incidences are expressed per million urban people exposed.

The aggregate risk from mobile sources in the Six Month Study is 2.65 per million. In this study, the aggregate risk ranges from 1.80 to 10.58 per urban million. As seen in Table S-2, a few of the unit risk estimates have increased considerably since the release of the Six Month Study, most notably formaldehyde and 1,3-butadiene. If the formaldehyde risk in the Six Month Study is increased to reflect the updated unit risk, the aggregate risk in the Six Month Study would

Table S-2

Comparison of Results

	Composite Emission Factor (g/mile)		Unit Risk[a]		Annual Mobile Source Cancer Risk (per urban million)	
Pollutant	6 Month	This Study	6 Month	This Study	6 Month	This Study[b]
Formaldehyde	0.033	0.042-0.045	6.1×10^{-6}	1.3×10^{-5}	0.07	0.22-0.70
Benzene	0.131	0.100	6.9×10^{-6}	8.0×10^{-6}	0.31	0.44-1.0
PIC[c]	1.66×10^{-5}	ND[d]	4.2×10^{-1}	ND	2.07	ND
B(a)P	1.66×10^{-5}	e	3.3×10^{-3}	3.3×10^{-3}	0.02	e
Diesel Particulate	ND	e	ND	$0.20\text{-}1.0 \times 10^{-4}$	ND	0.76-3.67
Gasoline PIC/POM	ND	e	ND	e	ND	0.007-0.76
Gasoline Vapors			7.5×10^{-7}	1.18×10^{-6}	0.15	0.36
1,3-Butadiene	1.0×10^{-3}	0.024-0.026	4.6×10^{-7}	2.8×10^{-4}	0	0-3.16
Ethylene	ND	0.237-0.261	ND	2.7×10^{-6}	ND	0-0.29
Asbestos	ND	$4\text{-}28 \times 10^{-6}$	ND	$6.6\text{-}260 \times 10^{-4}$	ND	0.002-0.63
Cadmium	8.5×10^{-6}	1.9×10^{-6}	2.3×10^{-3}	1.8×10^{-3}	0.01	0.001
Ethylene Dibromide	1.3×10^{-4}	7.1×10^{-5}	5.1×10^{-4}	5.1×10^{-4}	0.02	0.01
				Total:	2.65	1.80-10.58

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[a] The risk estimates are 95% upper confidence limits.

[b] An urban population of 180 million was used.

[c] Diesel particulate and particle-associated organics from gasoline-fueled vehicles were considered to represent PIC in this study. Therefore, a PIC risk was not determined in this study.

[d] ND=Not Determined

[e] See text for explanation.

increase to 2.73 per million. If the 1,3-butadiene risk in the Six Month Study is increased to reflect the updated unit risk, the risk from 1,3-butadiene would no longer be negligible.

The Six Month Study did not explicitly include diesel or gasoline-fueled particulate or gas phase exhaust organics (with the exception of formaldehyde and benzene). It did include a broad category of pollutants referred to as products of incomplete combustion (PIC). PIC are postulated to be primarily polynuclear aromatic hydrocarbons. PIC therefore includes most of the organics on motor vehicle particulate and some gas phase exhaust organics. The unit risk for PIC was derived from epidemiological studies of the general population and studies of occupational exposure to PIC (e.g., coke oven emissions). The PIC unit risk is expressed per unit of exposure of benzo(a)pyrene (B(a)P).

In the Six Month Study, the B(a)P emission factor for mobile sources was used to calculate an annual average exposure. The B(a)P exposure was then multiplied by the PIC unit risk to estimate annual cancer incidence due to PIC from mobile sources. The same process was performed for all other sources emitting B(a)P to determine cumulative annual cancer incidence from PIC. Since roughly 75 percent of the total B(a)P emissions were said to be attributable to mobile sources, 75 percent of the total PIC cancer incidence is due to mobile sources. The PIC risk from mobile sources in the Six Month Study is estimated to be 2.07 per urban million.

In this study, risks from diesel particulate and gasoline particle-associated organics (referred to as gasoline PIC/POM where POM is polycyclic organic matter) were analyzed separately. The sum of the diesel particulate and gasoline PIC/POM risks will be considered to represent PIC. Using this approach, the total PIC risk ranges from 0.77-4.43 per urban million. The PIC risk obtained in the Six Month Study lies within this range.

When reviewing the results of this study, the following are a few of the important limitations that should be considered. The report only accounts for a small number of the mobile source pollutants known to be emitted, and does not consider reactions of mobile source pollutants in the atmosphere. Resulting secondary pollutants may be more or less carcinogenic than what was originally emitted. The report also does not fully and accurately take into account seasonal variations in emissions. This may result in underestimating the risk since it is likely some air toxics emissions will increase with decreasing temperature. The emission factors were developed using 75°F as the ambient temperature. The risks presented are assumed to be additive. The risk projections for 1995 are based on the emission standards currently in place. Changes in fuel composition are not considered. The risk projections do not consider use of alternative fuels, such as methanol. Neither do the current or projected risk estimates consider use of alcohol/gasoline blends.

The following is a brief summary of each pollutant/ pollutant category covered in this report. It includes current research and regulatory activity. It should be read when reviewing the tables, since it describes important uncertainties. Literature references are not given in this summary, but are in the body of the report.

Diesel Particulate - Diesel particulate exhaust is composed of an elemental carbon core with hundreds of adsorbed organic compounds ranging from C_{14} to about C_{40}. Over 90 percent of diesel particulate is less than 1 micron in size. The light-duty vehicle and truck emissions standard is currently 0.6 gram/mile. New standards, effective in 1987, are 0.20 gram/mile for light-duty vehicles and 0.26 gram/mile for light-duty trucks. For heavy-duty diesel engines, there is currently no diesel particulate emission standard. A standard of 0.6 gram/brake horsepower-hour (g/bhp-hr) begins in 1988, with increasingly more stringent standards effective in 1991 and 1994. These increasingly stringent standards are accounted for in the 1995 projections.

Diesel particulate was found to be mutagenic in the late 1970's. Subsequent studies revealed that nitropolynuclear aromatic hydrocarbons (nitro-PAH), specifically, nitropyrenes, dinitropyrenes and nitrohydroxypyrenes together account for much of the mutagenicity observed. The organics extracted from diesel particulate and other known carcinogens such as coke oven emissions were tested in a battery of bioassays. These included bacteria and mammalian cell bioassays, and one skin painting study with SENCAR mice. Animal inhalation studies were conducted at that time but gave negative or inconclusive results. The unit risk for diesel particulate was determined by comparing the potency of diesel particulate with the potencies of the other carcinogens determined in these tests. The range of upper confidence limit unit risks used in this study, $0.2\text{-}1.0\times10^{-4}$, is based on various analyses of the comparative potency data. Several animal inhalation experiments have been recently completed which, in contrast to the earlier studies, show that diesel exhaust causes lung tumors in rats. After being analyzed, these experiments may help narrow the range of unit risks.

Total diesel particulate emissions in 1986 were estimated to be 274,000 metric tons, or roughly 3.9 percent of the total suspended particulate (TSP) emissions. Diesel particulate emissions are projected to drop to 125,000-154,000 metric tons/year in 1995. This is due to the more stringent diesel particulate standards. The range of emissions estimates in 1995 is due to the range of diesel sales assumed.

The annual mean exposure level, estimated using a modified version of the NAAQS exposure model (NEM) for CO, was 2.6 ug/m^3 in 1986. This exposure level accounts for both indoor and outdoor exposure to diesel particulate. This represents roughly 5 percent of the 1984 annual geometric mean TSP concentration. This level drops to 1.2-1.6 ug/m^3 in 1995. The resulting annual lung cancer risk from diesel particulate exposure for the U.S. population is 178-860 in 1986 and 92-443 in 1995.

Formaldehyde - Formaldehyde is emitted in the exhaust of both gasoline- and diesel-fueled vehicles. It has the chemical formula CH_2O. Formaldehyde is of interest due to its photochemical reactivity in ozone formation and suspected carcinogenicity. An upper confidence limit unit risk of 1.3×10^{-5} was used. It is based on a single study in which rats exposed to formaldehyde developed malignant and benign tumors in the nasal cavities; the unit risk is based on malignant tumor formation only. The current consensus in EPA favors use of malignant tumors only. EPA's Office of Toxic Substances is currently using the unit risk based on malignant tumors only in assessing the risk to garment workers and home residents. The consideration of benign tumors would increase the formaldehyde risk presented in this report by a factor of 15.

Formaldehyde exhaust emissions from motor vehicles correlate well with exhaust hydrocarbon (HC) emissions. For this analysis, formaldehyde emissions were expressed as a weight percentage of exhaust HC. These percentages were then applied to the exhaust HC output from the MOBILE3 emissions model for 1986 and 1995 to obtain the formaldehyde emission factors. In this way, deterioration and other effects are included. The percentages generally vary from 1 to 4 percent, depending on the vehicle class. Mobile source formaldehyde emissions in 1986 were estimated to be roughly 71,000 metric tons, or roughly 28 percent of the total formaldehyde emissions in the U.S. Mobile source formaldehyde emissions are expected to drop to 41,000 metric tons in 1995. This is due to the increasing use of 3-way and 3-way plus oxidation catalyst-equipped gasoline-fueled vehicles together with the phase out of non-catalyst-equipped vehicles. The result is a marked decrease in projected HC and, by association, formaldehyde emissions.

Nationwide exposure levels, using the modified CO NEM, are roughly 1.04-1.13 ug/m^3 and 0.59-0.65 ug/m^3 for 1986 and 1995, respectively. The resulting risk is 46-50 in 1986 and 29-31 in 1995. The range accounts for both the presence and absence of an Inspection/Maintenance program. This attempts to account for direct emissions only and does not account for either the destruction or photochemical formation of formaldehyde in the atmosphere.

Another approach intended to include photochemistry was also developed. With this approach, a mobile source fraction was applied to an annual average formaldehyde concentration developed by OAQPS using available ambient monitoring data. Mobile sources account for 28 percent of the total VOC emissions and 30 percent of the formaldehyde emitted directly. Assuming that the VOC from all sources have the equivalent potential to form formaldehyde, a mobile source fraction of 0.30 was selected. This fraction was applied to an urban population weighted average of 12.71 ug/m^3 (based on data obtained in 4 cities) and a rural concentration of 1.50 ug/m^3. Since the summer concentrations used to calculate the urban concentration probably represent maximum rather than average values, the risk estimates can be used to represent a plausible upper limit. Using this approach, the annual cancer risk from mobile source formaldehyde is 131 in 1986 and 77 in 1995. Combining both approaches, the cancer incidences due to mobile sources range from 46-131 in 1986 and 29-77 in 1995.

Research activity is planned or underway in three areas: 1) emissions characterization, 2) photochemistry, specifically, factors affecting formaldehyde formation in the atmosphere, and 3) ambient monitoring.

Benzene - Benzene is an aromatic hydrocarbon with the formula C_6H_6. It is present in both exhaust and evaporative emissions. Several epidemiologic studies have associated benzene with an increased incidence of leukemia. The upper confidence limit unit risk estimate determined from these studies is 8.0×10^{-6}.

Mobile sources (including refueling emissions) dominate the nationwide benzene emission inventory. In 1982, mobile source benzene emissions were roughly 250,000 metric tons, or 85 percent of the total benzene emissions. Of the mobile source contribution, 70 percent comes from exhaust, 14 percent from evaporative emissions and 1 percent from motor vehicle refueling.

Nationwide exposure levels from exhaust and evaporative emissions were estimated using the modified CO NEM. Benzene emissions were expressed as percentages of exhaust and evaporative HC. Percentages of exhaust HC vary from 1.1-5.12 percent for the vehicle classes; for evaporative emissions, the percentage varies from 0.35-1.53 percent. Nationwide exposure levels from both exhaust and evaporative emissions are roughly 3.1-3.2 ug/m^3 and 1.7-1.8 ug/m^3 for 1985 and 1995, respectively. (A previous analysis was relied on which used 1985 as the base year. It was assumed in this report that the calendar year emission factors for 1985 and 1986 would not differ significantly.) This assumed the presence of a standard, minimum I/M program. The range is due to consideration of both a low and high range evaporative emissions estimate for light-duty gasoline-fueled vehicles. Annual cancer incidences from exhaust and evaporative emissions are estimated to be 84-89 in 1985 and 50-52 in 1995. The reason for this marked decrease is the decrease in projected HC in 1995 and, thus, benzene emissions.

Exposure to benzene during refueling includes self-service refueling, occupational exposure (service station attendants) and community exposure in an urban area. Exposure to self-service refueling and occupational exposure was determined by measuring benzene levels in the region of the face of a person refueling a vehicle tank. The exposure in a typical urban area was estimated using a dispersion model. Annual cancer incidences from benzene refueling are estimated to be 8 in 1985 and 7 in 1995. Total cancer incidences from benzene exhaust, evaporative and refueling emissions are 92-97 in 1985 and 57-59 in 1995.

An alternative approach, similar to that used for formaldehyde, was also developed. With this approach, a mobile source fraction was applied to estimated urban and rural concentrations developed using available ambient monitoring data. Mobile sources account for 85 percent of the total benzene emissions. Therefore, a fraction of 0.85 was applied to an urban population weighted average of 10.24 ug/m^3 and a rural concentration of 7.52 ug/m^3. Using this approach, the annual lung cancer risk from mobile source benzene is 223 in 1985. Based on the NEM modeling, emissions of benzene from mobile sources are projected to decrease roughly 40 percent from 1985 to 1995. Accounting for this decrease and the projected population increase, the annual lung cancer risk is 145 in 1995. Combining both approaches, the lung cancer incidences due to mobile sources range from 92-223 in 1985 and 57-145 in 1995.

The California Air Resources Board (CARB) has also attempted to determine the risk posed by benzene emissions from various sources in 1984 and 2000. The vehicular contribution is estimated to be 21-166 cancer cases in California in 1984 and 20-154 cancer cases in 2000. Expressed as individual risk, the vehicular contribution is $8.1\text{-}64.3\times10^{-7}$ cancers/person in 1984 and $6.4\text{-}49.0\times10^{-7}$ cancers/person in 2000. In this case, the range is due to a range of unit risks used by CARB. The range of risk estimates given in this report, when expressed as individual risk, are $3.8\text{-}9.3\times10^{-7}$ cancers/year in 1986 and $2.2\text{-}5.6\times10^{-7}$ cancers/year in 1995. The upper bound of these risk estimates are roughly equivalent to the lower bound of the CARB estimates because, for these estimates, the ambient concentrations and unit risks are roughly equivalent. The upper bound CARB estimates use an upper bound unit risk that is roughly 7 times higher than the EPA-CAG unit risk used in this report.

The California Air Resources Board (CARB) is considering implementing regulations requiring control of motor vehicle benzene emissions. OAQPS has designated benzene as a hazardous air pollutant under Section 112 of the Clean Air Act and is implementing necessary controls for stationary sources. OMS is including benzene impacts in its assessment of VOC control options.

Gasoline Vapors - Totally vaporized gasoline has been found to cause a statistically significant increase in kidney tumors in male rats and liver tumors in female mice. An upper confidence limit based on the rat data is 1.18×10^{-6}.

Exposure to gasoline vapors during refueling was estimated based on an American Petroleum Institute (API) study that involved measuring gasoline and vapor levels in the region of the face of a person refueling a vehicle tank. The exposure in a typical urban area for these refueling emissions was also estimated using the Industrial Source Complex dispersion model to calculate annual concentrations. Based on these exposures, the risk from gasoline vapors (excluding benzene) was estimated as 65 lung cancer incidences per year.

EPA has not made a decision on what controls should be proposed for gasoline vapors.

Gas Phase Organics - Gas phase organics, or volatile organic compounds (VOC), are present in both exhaust and evaporative emissions. Over 300 VOC have been identified. The majority of VOC consist of unsaturated and saturated hydrocarbons along with benzene, alkyl benzenes, aliphatic aldehydes and a variety of polycyclic aromatic hydrocarbons. Most of the known mutagenicity of motor vehicle emissions is associated with the particulate phase, however.

Of all the VOC emitted from motor vehicles, only benzene, formaldehyde, benzo(a)pyrene (B(a)P), ethylene, and 1,3-butadiene have unit risks. Gas phase B(a)P was considered with particle-associated B(a)P since the majority of B(a)P is in the particulate phase. Ethylene emissions are present in vehicle exhaust and constitute from 6 to 13 percent of the exhaust HC emissions. Exposures were estimated using the modified NEM model. Based on the upper confidence limit unit risk provided in the Six Month Study (2.7×10^{-6}), risk estimates of lung cancer incidence for 1986 and 1995 are 55-60 and 29-31, respectively. The range accounts for the presence and absence of an I/M program. The unit risk is extremely tentative, however, since there is no available direct evidence that ethylene is carcinogenic. The unit risk for ethylene was estimated based on assumptions regarding its potency relative to ethylene oxide, a metabolite of ethylene and an animal carcinogen. For this reason, a lower risk estimate of zero is used.

1,3-Butadiene is a photochemically reactive compound present in vehicle exhaust. The Six Month Study found no risk associated with emissions of 1,3-butadiene from mobile sources; however, since the release of the Six Month Study the unit risk has increased by a factor of 1000. Updated emission factors for 1,3-butadiene also appear higher but determination of an accurate emission factor is difficult because 1,3-butadiene and n-butane coelute and thus have the same retention point on the gas chromatograph. Emission characterization studies to date have not attempted to determine the percentage of the peak due to 1,3-butadiene. Therefore, assumptions must be made about the percentage each compound contributes to this peak. It was assumed in this report that 15 percent of the peak was due to 1,3-butadiene, based on data collected in New York's Lincoln Tunnel, although ambient data indicate that the actual percentage could well be much lower. Fifteen percent was chosen as an upper limit. Based on data from in-use gasoline-fueled vehicles, 1,3-butadiene is roughly 0.94 percent of the total exhaust HC as measured by the flame ionization detector (FID). Due to the lack of data for the other vehicle classes, this percentage was simply applied to the MOBILE3 composite exhaust HC emission factor. It was further assumed that the percentage would remain the same from 1986 to 1995.

The modified NEM was used to estimate exposures. Nationwide urban exposure in 1986 is estimated to be 0.60-0.66 ug/m^3. The range accounts for the presence and absence of an I/M program. These exposure estimates are for direct emissions of 1,3-butadiene and do not account for reactions of 1,3-butadiene in the atmosphere. Available ambient monitoring data were reviewed and compared to the exposure estimates. Average mean values in urban settings range from 0.24-24.23 ug/m^3, although the accuracy of the analytical methods used is uncertain. The NEM urban exposure estimate lies within this range.

Using the exposure estimates in conjunction with the upper confidence limit unit risk estimate ($2.8\text{x}10^{-4}$), estimates of lung cancer incidence for 1986 and 1995 are 593-656 and 391-460, respectively. Preliminary emission characterization results indicate the presence of 1,3-butadiene, but the amount has not yet been quantified. Therefore, a lower risk estimate of zero will also be used. The resulting ranges of cancer incidences for 1986 and 1995 given in Table S-1 are 0-656 and 0-460, respectively. OAQPS is currently working on a source assessment document for 1,3-butadiene which should be completed sometime in late 1987.

The atmospheric photochemical reaction products of mobile source VOC are largely unknown. Smog chamber experiments have produced unidentified gas phase mutagens starting with simple VOC compounds that are present in vehicle emissions. It appears that, as non-catalyst-equipped vehicles are phased out of the fleet, the reactivity of vehicle exhaust will decrease. This is evidenced by the decreasing percentages of reactive olefins and aromatics, coupled with the increasing percentage of less reactive paraffins (particularly methane) in the exhaust. The Integrated Air Cancer Project (IACP) is a long-term effort by EPA ORD to identify the principal airborne carcinogens and their sources, and may clarify the risk posed by reaction products.

Organics Associated with Non-Diesel Particulate - Gasoline-fueled vehicles emit far less particulate than their diesel counterparts. It is thought that a number of nitro-polycyclic aromatic hydrocarbon (PAH) compounds account for much of the mutagenicity of diesel particulate emissions. Particulate emissions from gasoline-fueled vehicles contain significantly less of these nitro-PAH's; however, the mutagenicity of the gasoline soluble organic fraction (SOF), expressed as revertants/ug SOF, is greater than diesel SOF. Also, unlike diesel SOF, the mutagenic activity of gasoline SOF increases with the addition of S9 activation, indicating indirect-acting activity. This suggests that the classical PAH's may be responsible for the mutagenicity of gasoline SOF, rather than the nitro-PAH's.

The organics associated with gasoline particulate were considered to represent gasoline products of incomplete combustion (PIC). Three different approaches were taken to estimate the risk from gasoline PIC. The first approach assumes the risk from B(a)P emissions adequately represents the risk of all gasoline PIC. B(a)P is emitted primarily from gasoline-fueled vehicles. The annual cancer risk of B(a)P from gasoline-fueled vehicles was determined by adjusting the upper confidence limit B(a)P risk obtained in the Six Month Study to account for the difference in emission factors. The resulting annual cancer risk in 1986 is 0.007 per urban million, or 1.3 cancer incidences, assuming an urban population of 180 million. B(a)P emissions should decrease with the phase-out of leaded fuel. In 1995, the cancer risk is projected to decrease to 0.004 per urban million, or 0.78 cancer incidences, assuming an urban population of 195 million.

The second approach uses B(a)P emission factors from gasoline-fueled vehicles together with the PIC upper confidence limit unit risk used in the Six Month Study (which is expressed per unit of exposure of B(a)P) to estimate the annual cancer risk of PIC from gasoline-fueled vehicles. This approach assumes that B(a)P is an adequate surrogate for all PIC compounds. It should be noted that this approach is rather uncertain as the mix of PIC compounds differs among source types. The annual cancer risk was determined by adjusting the PIC risk obtained in the Six Month Study to account for the difference in B(a)P emission factors. The resulting annual cancer risk is 0.68 per urban million, or 122 cancer incidences in 1986. In 1995, the cancer risk is projected to decrease to 0.37 per urban million, or 72 cancer incidences.

The third approach uses estimated emission rates of gasoline particle-associated organics (as an unspeciated mixture) together with an upper confidence limit unit risk for these organics. Exposures were estimated using the modified NEM model.

Estimated composite emission factors for gasoline particle-associated organics, with and without an I/M program, are 0.0075-0.0082 g/mile in 1986 and 0.0048-0.0058 g/mile in 1995. Total organic emissions are not projected to decrease as much as B(a)P emissions. This is because emission data indicate that use of a catalyst reduces B(a)P emissions to a greater extent than total organic emissions.

A unit risk estimate for gasoline particle-associated organics has been estimated, based on data from the only catalyst-equipped vehicle tested for particle organic mutagenicity. The vehicle had exceptionally high exhaust emissions, comparable to those from a non-catalyst equipped vehicle. It was originally chosen for testing in 1979 on this basis since it was easier to collect enough extractable organics for analysis. The mutagenic activity of the particle-associated organics from this vehicle, as indicated by the Ames Salmonella strain TA-98 bioassay, is on the low end of the range, when compared with other catalyst-equipped vehicles. As a result, the vehicle should be considered to be of uncertain representativeness. An upper confidence limit unit risk estimate based on this vehicle is $2.5x10^{-4}$.

The total risk in 1986, accounting for both urban and rural exposure, is 163-176 cancer incidences and drops to 115-136 cancer incidences in 1995.

For this report, a range of risk estimates for gasoline PIC will be reported, which encompasses the results of all three approaches. The resulting range of cancer incidences is 1.3-176 in 1986 and 0.78-136 in 1995.

Dioxins - The major dioxin compound of interest and the one considered in this report is 2,3,7,8-tetrachlorodibenzo-p-dioxin. This dioxin compound exists in the particulate state or is adsorbed onto particulates. Some qualitative analytical measurements have found dioxin to be present in the muffler scrapings of vehicles using either leaded or unleaded gasoline. It appears to be emitted in only trace quantities (e.g., 10^{-9} g/mile) in vehicle exhaust. OAQPS plans to make a decision within the coming year on whether to list dioxin as a hazardous air pollutant.

Asbestos - Asbestos is used in brake linings, clutch facings and automatic transmissions. About 22 percent of the total asbestos used in the U.S. in 1984 was used in motor vehicles. Asbestos emissions from vehicles with front disc brakes and rear drum brakes ranged from 4-28 ug/mile. Based on these emission rates, maximum annual average asbestos levels in urban areas due to motor vehicles are estimated to range from 0.25 to 1.75 nanograms per cubic meter (ng/m^3). Asbestos from mobile sources appears to account for roughly 1-10 percent of urban asbestos concentrations, although mobile sources could be responsible for as much as 70 percent, under worst case emissions conditions.

EPA has not attempted to quantify the excess mortality from asbestos exposure. The National Academy of Sciences (NAS) has, however, estimated life time risks for persons in urban areas. Based on the data in the NAS report, the individual annual cancer risk from urban levels of asbestos is estimated to range from $9 \times 10^{-9} - 3.6 \times 10^{-7}$ per ng/m^3 exposure. Assuming an urban population of 180 million, mobile source asbestos emissions could be responsible for as many as 113 cancer incidences per year.

EPA's Office of Pesticides and Toxic Substances (OPTS) has proposed regulations under Section 6 of TSCA to ban certain uses of asbestos and to allocate permits to mine and import asbestos which would restrict its remaining uses. EPA is also considering a ban on asbestos friction products about 5 years after the final rules are promulgated. The fact that there are not yet available good substitutes to replace asbestos in certain automobile and truck brakes, however, may push back EPA's goal for banning and phasing out asbestos for such uses.

Vehicle Interior Emissions - A total of 147 compounds have been identified in vehicle interiors. In closed vehicles under high temperatures, vinyl chloride was present at levels ranging from below 2 ppb to 7 ppb. A number of other carcinogenic compounds were identified qualitatively. Due to the low exposure level, no significant risk should be present from vehicle interior emissions.

Ethylene Dibromide (EDB) and Cadmium - Updated emission factors for EDB and cadmium were estimated. The risk estimates for EDB and cadmium were then determined by adjusting the risk obtained in the Six Month Study to account for the difference in emission factors. As seen in Tables S-1 and S-2, the risks from these pollutants are negligible in comparison to other mobile source pollutants.

1.0 INTRODUCTION

Considerable effort is underway within the Environmental Protection Agency (EPA) to determine the magnitude of the air toxics problem in the United States. For the purposes of this report, air toxics are defined as carcinogens associated with air pollution. The overall objective is to develop quantitative estimates of the upper-bound cancer risk associated with air toxics emissions, and to determine the relative contribution of specific pollutants and sources.

In 1985, EPA completed a report which is commonly referred to as the Six Month Study. This study contains estimated cancer risks due to 15 to 45 toxic air pollutants (the number of pollutants examined varied with the three different analyses that were performed).[1]* The estimates of cancer incidence from these analyses ranged from 1300 to 1700 cases annually nationwide for all pollutants combined. These estimates were based on use of upper-bound unit risks. The analyses further indicated that mobile sources may be responsible for a large portion of the aggregate cancer incidence. The Six Month Study, however, was broad in nature with the goal to obtain a quick assessment of the air toxics problem in the United States and to guide further studies.

The purpose of this report is to focus on air toxics emissions from mobile sources. Specific pollutants or pollutant categories which will be discussed include diesel particulate, formaldehyde, benzene, gasoline vapors, gas phase organics, organics associated with non-diesel particulate, dioxins, asbestos, vehicle interior emissions, and metals. For each pollutant or pollutant category, available information is given on the formation and composition, control technology, emissions, ambient concentrations, unit risk estimate, the current and projected public health impact (or risk), and current EPA regulatory and/or research activity.

The results obtained are then compared to the earlier EPA report.[1] Following this is a summary of the total risk from mobile source air toxics emissions, together with the limitations inherent in the estimate.

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* Numbers in brackets designate references at the end of the report.

2.0 DIESEL PARTICULATE

It should be noted that, since there has not been a recent analysis by EPA of diesel particulate emission factors and exposure, a detailed analysis was performed during preparation of this report. Other pollutants are not treated as thoroughly.

2.1 Formation and Composition

Particulate emissions from diesel-equipped vehicles are formed as a result of incomplete combustion of the fuel. The particles in diesel exhaust differ both in quantity and composition from particles in gasoline engine exhaust. Diesel-equipped vehicles can emit from 30 to 100 times more particulate mass, on a grams per mile basis, than gasoline-powered, catalyst-equipped vehicles.[2] Over 90 percent of diesel particulates are less than 1 micron in size and are therefore small enough to be inhaled and deposited deep within the lungs.[3] Virtually all are less than 10 microns.

Gasoline particulate emissions from catalyst-equipped vehicles using unleaded fuel are very low in mass and consist largely of sulfates and also some organics. Diesel particulate emissions, on the other hand, are very complex, being composed of carbonaceous matter with condensed and/or absorbed fuel and lubricant components and other varied combustion products. The soluble organic fraction (SOF) of the total diesel particulate mass (i.e., organics extractable with methylene chloride) varies significantly. Typically, diesel passenger car SOF ranges between 5 and 50 percent of the total particulate mass.[4]

The chemical composition of diesel SOF is complex. Generally, diesel SOF ranges from C_{14} to about C_{40}.[4] Hundreds of compounds are present and the analytical capability does not exist to identify every compound. Instead, effort has been focused to identify the chemical classes and specific compounds associated with the SOF that are mutagenic in the Ames bioassay test.

The mutagenicity of diesel SOF decreases upon the addition of S9 activation, indicating direct-acting frameshift mutagenic activity. The SOF was solvent-partitioned into organic acids, bases, and neutral components; the neutral components were further fractionated and the mutagenic activity of each fraction was determined using the Ames Salmonella typhimurium/microsome assay. The moderately and highly polar neutral compounds account for 89-94 percent of the mutagenic activity and only 32 percent of the mass.[5] Gas chromatography/mass spectroscopy identified nonmutagenic fluorenones and methylated fluorenones as major constituents of the moderately and highly polar neutral fractions but they did not account for the direct-acting activity observed. Studies with nitroreductase-deficient strains of Salmonella typhimurium suggested that nitrated compounds are present. Subsequent studies show that nitropyrenes, dinitropyrenes and nitrohydroxypyrenes together account for much of the mutagenicity observed.[5, 6]

EPA conducted a large research program to evaluate the health effects associated with exposure to diesel emissions, with particular emphasis on the organic extracts. The culmination of this effort was an estimation of the unit risk* for diesel particulate.[7]

2.2 Control Technology

A variety of approaches are being developed by manufacturers to control diesel particulate emissions. One approach is the use of a catalytic converter to oxidize and remove the soluble organics absorbed on the particulate. The other, more popular approach is the use of a particulate filter or trap. Reduction of particulate formation via combustion chamber modifications is also being investigated. The primary traps being evaluated are: 1) a catalyzed ceramic monolith trap, 2) a ceramic monolith trap, and 3) a catalyst-coated wire mesh trap. These traps are being evaluated for both light-duty and heavy-duty applications.

For light-duty applications, Daimler-Benz introduced in the 1985 model year a catalyzed ceramic monolith trap in some of its turbocharged diesels. Most manufacturers, however, are investigating the feasibility of non-catalyzed, ceramic monolith traps.

A trap must be periodically regenerated by oxidizing the collected particulates. Otherwise, particulates collected on a trap can cause the exhaust back pressure to increase and adversely affect fuel economy and vehicle performance. Active and passive regeneration techniques have been assessed. An active technique would use a diesel-fueled burner or electric resistance heater to raise the temperature of the engine exhaust gas flowing into the trap or to raise the temperature of the trap itself to the ignition temperature of the collected particulates, thus combusting the particulate which regenerates the trap. It should be noted that the organic emissions from trap regeneration have not been fully characterized.

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*Unit risk is defined as the individual life time excess cancer risk from continuous exposure to 1 ug carcinogen per m^3 inhaled air. Assuming a life time is 70 years, the excess lung cancer risk in 1 year is derived by simply dividing the unit risk by 70. Using this approach, latency is ignored.

A passive technique uses catalytic material to lower the ignition temperature of the collected particulate. One method is to apply the catalytic coating to the trap substrate itself. Daimler-Benz introduced in the 1985 model year a catalyzed ceramic monolith trap in some of its turbocharged diesels. Johnson-Matthey, a catalyst manufacturer involved in trap development, is investigating the merits of a catalyst-coated stainless steel wire mesh trap.

The other passive method is the use of a metal fuel additive in conjunction with a ceramic wall flow monolith trap. This yields the lowest ignition temperature of any other passive method. The most promising metal fuel additives to date are manganese and copper compounds.

2.3 Emissions

In this section, the Federal diesel particulate standards are presented. This is followed by an estimation of in-use emission factors by model year for each vehicle class (i.e., light-duty car, light-duty truck and heavy-duty vehicle). These model year emission factors are then used in conjunction with information on diesel sales fractions and fraction of diesel travel by model year to obtain calendar year emission factors for each vehicle class. For this analysis, calendar years 1986 and 1995 were selected. Nationwide diesel particulate emissions (metric tons/year) for 1986 and 1995 are then calculated by combining the emission factors with projected vehicle miles traveled (VMT) data for 1986 and 1995. The results are then compared to the most recent national particulate emission estimate.

2.3.1 Emission Standards

Table 2-1 provides a summary of the Federal diesel particulate emission standards for light-duty cars and trucks and heavy-duty engines. The test procedure specified for light-duty vehicles is the CVS-75 or FTP procedure, a constant volume sample test which includes both cold and hot starts. For heavy-duty vehicles, a transient test procedure is currently used. This is an engine dynamometer procedure with starts, stops, and speed/load changes. In order to meet the future Federal standards (1987 and later), some light-duty and heavy-duty vehicles will need to be equipped with a particulate aftertreatment system, such as a trap.

Table 2-1

Diesel Particulate Standards

Light-Duty Vehicles

Year	Standard (grams per mile) Cars	Standard (grams per mile) Trucks
1981 and prior	-	-
1982-1986	0.60	0.60
1987 and later	0.20*	0.26*

Heavy-Duty Engines

Year	Standard (grams per brake horsepower-hour)
1987 and prior	-
1988-1990	0.60
1991-1993	0.25** 0.10***
1994 and later	0.10**

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* Emissions averaging may be used to meet this standard. For trucks, some restrictions apply.

** Emissions averaging may be used to meet this standard. Some restrictions apply.

*** For urban bus engines, the standard is 0.10 g/Bhp-hr beginning in 1991. Particulate averaging is not allowed with this standard.

NOTE: In an emissions averaging program, the manufacturer determines emission limits for each vehicle/engine family. Family emission limits are allowed to exceed the standards; however, the weighted average of the family emission limits myst be in compliance with the applicable standard. For cars and trucks, the average emission level is based upon a production-weighted average of the family emission limits. For heavy-duty engines, the average emission level is determined by calculating a production- and horsepower-weighted average of the family emission limits.

2.3.2 Emission Factors by Model Year

The light-duty and heavy-duty diesel particulate emission factors are given in Table 2-2. The heavy-duty sub-groups are defined by gross vehicle weight rating (GVWR) as follows:

Class 2B = 8,500 to 10,000 lbs.
Classes 3-5 = 10,001 to 19,500 lbs.
Classes 6 = 19,501 to 26,000 lbs.
Classes 7 and 8 = 26,001 lbs and up.

The emission factors for light-duty cars and trucks were obtained from an EPA report titled, "Diesel Particulate Study."[8] For 1987 and later, these emission factors are considered to be, for the purpose of this report, the respective emission standards. For heavy-duty vehicles, the emission factor assumed for 1983 and prior is 1.0 grams per brake horsepower-hour (g/Bhp-hr). For 1984-1987, the emission factor assumed is 0.75 g/Bhg-hr. This is based on transient test data for a variety of in-use heavy-duty vehicles.[9] For 1988-1993, the emission factor assumed is the standard. For 1994 and 1995, the standard was adjusted upward slightly to account for projected trap failure. Within classes 7-8, the emission factor for buses was adjusted upward to account for projected trap failure beginning in 1991. The equation used to make this adjustment is given and discussed in detail in reference 8.

It was necessary to convert the heavy-duty emission factors from g/Bhp-hr to g/mile. As seen in Table 2-2, the g/mile emission factors for the heavy-duty subgroups differ even though they are subject to the same g/Bhp-hr standard for 1988 and later and similar in-use g/Bhp-hr emissions prior to 1988. The conversion of g/Bhp-hr to grams/mile is dependent on the engine brake-specific fuel consumption (BSFC, lb/Bhp-hr), the fuel density (lb/gallon) and the fuel economy (mile/gallon) of the particular vehicle/engine configuration. Since the BSFC and fuel economy varies among the different heavy-duty subgroups, estimated g/mile emissions vary as well. Conversion factors for the heavy-duty subgroups by model year were based on the information provided in reference 10. These conversion factors are consistent with those used in the MOBILE3 emissions model.[11]

2.3.3 Emission Factors for Calendar Years 1986 and 1995

The next step is to combine the model year emission factors for each vehicle type into a single, weighted calendar year emission factor for each vehicle type. Model year data for the previous 20 years are used, i.e., for 1986, model year data back to 1967 are used; for 1995, model year data back to 1976 are used. Each model year's emission factor is multiplied

Table 2-2

Diesel Particulate Emission Factors

Light-Duty Vehicles
grams per mile

Model Year	Cars	Trucks
1978 and prior	0.70	0.90
1979	0.80	0.90
1980	0.50	0.50
1981-1986	0.27	0.28
1987 and later	0.20	0.26

Heavy-Duty Engines*

grams per mile

Model Year	2B	3-5	6	7-8
1967	1.00	1.71	1.71	2.99
1968	1.00	1.71	1.73	3.02
1969	1.00	1.71	1.75	3.06
1970	1.00	1.71	1.75	3.09
1971	1.00	1.71	1.79	3.14
1972	1.00	1.71	1.82	3.20
1973	1.00	1.71	1.84	3.24
1974	1.00	1.71	1.85	3.29
1975	1.00	1.71	1.86	3.33
1976	1.00	1.71	1.87	3.24
1977	1.00	1.71	1.87	3.28
1978	1.00	1.71	1.86	3.23
1979	0.99	1.53	1.86	3.18
1980	0.98	1.34	1.86	3.16
1981	0.98	1.16	1.86	3.13
1982	0.97	0.97	1.87	3.09
1983	0.97	0.97	1.85	3.08
1984	0.73	0.73	1.37	2.30
1985	0.73	0.73	1.36	2.29
1986	0.73	0.73	1.34	2.28
1987	0.72	0.72	1.33	2.28
1988	0.58	0.58	1.06	1.81
1989	0.57	0.57	1.06	1.79
1990	0.57	0.57	1.06	1.77
1991	0.24	0.24	0.44	0.69
1992	0.24	0.24	0.44	0.69
1993	0.24	0.24	0.44	0.67
1994	0.09	0.09	0.19	0.30
1995	0.09	0.09	0.18	0.29

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* See text for explanation of the g/Bhp-hr emission factors used.

by that model year's fraction of calendar year VMT and the diesel sales fraction for that model year, and then summed across all 20 model years.

The fraction of travel by model year for the light-duty vehicles and trucks is given in Table 2-3. These travel fractions are dependent on vehicle age rather than model year; therefore, the same travel fractions were used to determine both the 1986 and 1995 emission factors. These travel fractions were obtained from the MOBILE3 emissions model.[11]

The fraction of travel by model year for the heavy-duty subclasses for calendar years 1986 and 1995 are given in Tables 2-4 and 2-5, respectively. For 1986, the travel fractions for heavy-duty gasoline-fueled trucks were used for subclasses 2B, 3-5 and 6. The travel fractions for heavy-duty diesels were used for subclasses 7-8, since this combined subclass is dominated by heavy-duty diesels. For 1995, the travel fractions for heavy-duty gasoline-fueled trucks were used for subclasses 2B and 3-5; travel fractions for heavy-duty diesels were used for subclasses 6 and 7-8. Unlike the heavy-duty gasoline-fueled trucks, the mileage distributions and resulting travel fractions for heavy-duty diesels are dependent on model year and reflect the increasing penetration of diesels in the lower mileage, lighter weight classes of the heavy-duty truck category. These travel fractions were calculated based on the information provided in Reference 11.

Projecting future diesel sales is rather uncertain. Light-duty diesel sales in particular have dropped off quite dramatically and should fall well short of projections made just a few years ago that light-duty diesels would account for 25 percent of light-duty vehicle sales in 1995. To account for these uncertainties, low and high diesel sales scenarios were developed. Both scenarios incorporate actual sales data to the extent available.

For light-duty vehicles, actual diesel sales fractions up to and including 1985 are available and were used.[12,13] For 1986 and beyond, the low diesel sales scenario assumes that the light-duty vehicle diesel sales fraction will remain constant at the 1985 level (0.009). The high diesel sales scenario uses MOBILE3 projections of diesel sales for 1995 (0.009 in 1986 with a gradual increase to 0.115 in 1995).

For light-duty trucks and heavy-duty subclass 2B, actual diesel sales fractions through 1984 were used.[12,13] For 1985 and beyond, the low diesel sales scenario assumes that the diesel sales fraction will remain constant at the 1984 level. The high diesel sales scenario uses the latest EPA/OMS projections for 1985-1995.[14]

Table 2-3

Fraction of Travel by Model Year for Light-Duty Vehicles and Trucks

Vehicle Age	LDV Travel Fraction	LDT Travel Fraction
1	.038	.035
2	.142	.129
3	.125	.114
4	.111	.101
5	.098	.088
6	.084	.078
7	.075	.067
8	.065	.058
9	.055	.050
10	.047	.043
11	.040	.037
12	.032	.031
13	.026	.026
14	.021	.022
15	.015	.018
16	.011	.015
17	.007	.012
18	.003	.009
19	.003	.006
20+	.004	.009

Table 2-4

Fraction of Travel by Model Year for Heavy-Duty Vehicles in Calendar Year 1986

Model Year	2B, 3-5 and 6	7-8
1986	.000	.000
1985	.227	.255
1984	.175	.186
1983	.134	.136
1982	.105	.105
1981	.080	.078
1980	.062	.057
1979	.049	.043
1978	.037	.033
1977	.028	.026
1976	.023	.019
1975	.017	.014
1974	.013	.011
1973	.010	.008
1972	.009	.006
1971	.006	.005
1970	.005	.004
1969	.004	.003
1968	.003	.002
1967	.013	.009

Table 2-5

Fraction of Travel by Model Year for Heavy-Duty Vehicles in Calendar Year 1995

Model Year	2B, 3-5	6	7-8
1995	.000	.000	.000
1994	.227	.292	.272
1993	.175	.208	.199
1992	.134	.146	.144
1991	.105	.105	.106
1990	.080	.074	.076
1989	.062	.052	.055
1988	.049	.037	.040
1987	.037	.026	.029
1986	.028	.019	.021
1985	.023	.013	.015
1984	.017	.008	.011
1983	.013	.006	.008
1982	.010	.004	.006
1981	.009	.002	.004
1980	.006	.002	.003
1979	.005	.001	.002
1978	.004	.001	.002
1977	.003	.001	.001
1976	.013	.003	.006

The diesel sales fractions for heavy-duty classes 3-5, 6 and 7-8 were based on actual data through 1985. For 1986 to 1995, the latest EPA/OMS projections were used.[14] These sales fractions were used for both the low and high diesel sales scenarios.

The diesel sales fractions for the light-duty vehicles and trucks are presented in Table 2-6. The diesel sales fractions for the heavy-duty vehicle classes are given in Table 2-7.

The resulting weighted emission factors for calendar years 1986 and 1995 are given below:

(g/mile)

			HDV			
	LDV	LDT	2B	3-5	6	7-8
Calendar Year 1986						
Low Diesel Sales	.008	.0105	.089	.040	.479	2.35
High Diesel Sales	.008	.0105	.091	.040	.479	2.35
Calendar Year 1995						
Low Diesel Sales	.003	.008	.061	.082	.330	.844
High Diesel Sales	.013	.041	.084	.082	.330	.844

2.3.4 Nationwide Diesel Particulate Emissions

In this section, nationwide diesel particulate emissions (metric tons/year) for 1986 and 1995 are calculated by combining the calendar year emission factors with projected vehicle miles traveled (VMT) data for 1986 and 1995.

The VMT fractions and resulting projected VMT for 1986 and 1995 is given in Table 2-8. These data were obtained from the MOBILE3 Fuel Consumption Model.[15] In this model, heavy-duty subgroups 2B and 3-5 were treated as one subgroup. To split the VMT fraction for these subgroups, data from EEA's Motor Fuel Consumption, Tenth Periodical Report were used.[16] Total VMT is projected to increase 19 percent from 1986 to 1995.

The projected nationwide diesel particulate emissions for 1986 and 1995 are given in Table 2-9. As seen in this table, nationwide diesel particulate emissions are projected to decrease roughly 44 to 54 percent from 1986 to 1995, despite the 19 percent projected increase in VMT. This is due to the rather small projected further infiltration of the fleet by diesels (with the exception of the heavier heavy-duty subclasses 7-8) together with increasingly stringent standards in future years.

Table 2-6

Low and High Diesel Sales Fractions by Model Year for Light-Duty Vehicles and Trucks

Model Year	Light-Duty Vehicles*		Light-Duty Trucks**	
	Low	High	Low	High
1995	.009	.115	.026	.339
1994	.009	.102	.026	.300
1993	.009	.089	.026	.263
1992	.009	.076	.026	.226
1991	.009	.063	.026	.188
1990	.009	.050	.026	.150
1989	.009	.044	.026	.119
1988	.009	.034	.026	.088
1987	.009	.019	.026	.057
1986	.009	.009	.026	.026
1985	.009		.026	.026
1984	.023		.026	
1983	.019		.042	
1982	.039		.092	
1981	.060		.082	
1980	.045		.048	
1979	.026		.015	
1978	.009		.010	
1977	.003		.005	
1976	.003		.003	
1975	.003		.002	
1974	.003		.000	
1973	.002		.000	
1972	.002		.000	
1971	.001		.000	
1970+	.000		.000	

---

*1970-1985 sales fractions are based on actual data and are the same for both scenarios. These fractions are not repeated in the high sales column in order to highlight the differences between scenarios.

*1970-1984 sales fractions are based on actual data and are the same for both scenarios.

Table 2-7

Low and High Diesel Sales Fractions by Model Year for Heavy-Duty Vehicles

Model	2B		3-5	6	7-8
Year	Low	High	Best	Best	Best
1995	.190	.300	.300	.712	.883
1994	.190	.300	.300	.693	.882
1993	.190	.300	.300	.674	.879
1992	.190	.300	.300	.655	.878
1991	.190	.286	.291	.635	.877
1990	.190	.273	.282	.616	.875
1989	.190	.259	.273	.597	.874
1988	.190	.245	.265	.578	.872
1987	.190	.231	.256	.559	.871
1986	.190	.218	.247	.540	.868
1985	.190	.204	.239	.521	.867
1984	.190		.000	.382	.849
1983	.153		.000	.312	.862
1982	.127		.000	.289	.845
1981	.000		.000	.322	.881
1980	.000		.000	.183	.871
1979	.000		.000	.114	.820
1978	.000		.000	.078	.815
1977	.000		.000	.070	.878
1976	.000		.000	.042	.823
1975	.000		.000	.032	.733
1974	.000		.000	.016	.770
1973	.000		.000	.016	.780
1972	.000		.000	.016	.760
1971	.000		.000	.015	.750
1970	.000		.000	.016	.750
1969	.000		.000	.000	.750
1968	.000		.000	.000	.750
1967	.000		.000	.000	.750

Table 2-8

VMT Fractions and VMT for 1986 and 1995

VMT Fractions

			HDV			
	LDV	LDT	2B	3-5	6	7-8
1986	.675	.225	.022	.004	.009	.065
1995	.671	.227	.025	.003	.006	.068

Projected VMT (10^9 miles)

			HDV				
	LDV	LDT	2B	3-5	6	7-8	TOTAL
1986	1098.53	365.25	35.16	6.11	15.34	106.48	1626.87
1995	1298.75	438.59	49.19	5.68	10.64	131.48	1934.33

Table 2-9

Nationwide Diesel Particulate Emissions (metric tons per year)

	1986		1995	
	Low Sales	High Sales	Low Sales	High Sales
LDV	8,788	8,788	3,896	16,884
LDT	3,835	3,835	3,509	17,982
HDV				
2B	3,129	3,200	3,001	4,132
3-5	244	244	466	466
6	7,348	7,348	3,511	3,511
7-8	250,228	250,228	110,969	110,969
TOTAL	273,572	273,643	125,352	153,944

2.3.5 Other Sources of Particulate

To put the preceding emission estimates into perspective the results are compared to national particulate emission estimates for 1984.[17] 1984 is the most recent year for which data are available.

Total particulate emissions in the U.S. in 1984 were projected to be 7.0 million metric tons. In comparison, diesel particulate emissions in 1986 were projected to be roughly 274,000 metric tons, or 3.9 percent of the total 1984 emissions. Transportation sources in 1984 were estimated to contribute 18.6 percent of the total, or 1.3 million metric tons. Diesel particulate emissions in turn currently appear to account for about 21 percent of the emissions from transportation sources.

2.4 Ambient Concentrations of Diesel Particulate

In this section, urban and rural concentrations of diesel particulate are estimated for 1986 and 1995, using a modified version of the EPA NAAQS Exposure Model (NEM) for CO. These concentrations are then compared to previous exposure estimates and to monitoring data for total suspended particulate.

2.4.1 Estimation of Urban and Rural Exposure

The model used provides an estimate of nationwide annual person-hours of exposure to any non-reactive mobile source pollutant of interest.[18] It is based upon the NEM developed originally by EPA's Office of Air Quality Planning and Standards (OAQPS). The NEM approach relies on an activity pattern model that simulates a set of population groups called cohorts as they go about their day-to-day activities. Each of these cohorts are assigned to a specific location type during each hour of the day. Each of several specific location types in the urban area are assigned a particular ambient pollutant concentration based on fixed site monitor data. The model computes the hourly exposures for each cohort and then sums up these values over the desired average time to arrive at average population exposure and exposure distributions. Annual averages are possible because a full year's data from fixed site monitors is an input to the model.

The NEM approach was designed to determine an integrated exposure to a pollutant by estimating both indoor and outdoor exposures to the pollutant. It was not designed to determine exposure from a particular source, such as mobile sources. Also, because its basic time unit is an hour, it does not account well for short periods spent in locations with high exposure such as an on-road vehicle. Hence, Southwest Research Institute (SwRI), under EPA contract, developed a new model based on the NEM for CO which could be used to better determine exposures resulting specifically from mobile source pollutants.[18] The CO NEM was used since outdoor CO is almost exclusively mobile source related. Since the CO monitor data, on which the CO NEM was based, can be assumed to be related to mobile source emission rates, exposure to other non-reactive mobile source pollutants can be modeled using this relationship.

The CO monitor data are used to provide CO concentration data for each neighborhood and most of the microenvironments. In each neighborhood or microenvironment, CO emission factors (in grams/minute) are chosen which are felt to best represent vehicle conditions in that neighborhood/microenvironment. For example, in an urban commercial neighborhood, an emission factor at 10 mph steady state is chosen since this is thought to best represent the vehicle conditions in this neighborhood. The emission factor is a fleet average emission factor, thus weighting emissions from both light-duty and heavy-duty vehicles. The model ratios the CO concentrations and appropriate CO emission factors for each neighborhood/microenvironment, so each neighborhood/microenvironment contains a ug/m^3/grams/minute factor. Emission factors in grams/minute for the pollutant of interest for each neighborhood/microenvironment are input to the model. The model simply multiplies the input emission factor (grams/minute) by the factor (ug/m^3/grams/minute) to obtain concentrations in each neighborhood/microenvironment for the pollutant of interest.

It should be noted that indoor concentrations (and therefore, exposure) due to ambient mobile source pollutants are also accounted for in the model. A scaling factor of 0.85 was applied to the appropriate neighborhood CO monitor data to estimate indoor exposures to the pollutant of interest in each neighborhood. The scaling factor was based on comparisons of indoor and outdoor CO levels of homes with no indoor CO sources (e.g., gas stove, smokers).

The model does not account for photochemical reactions. The exposure levels predicted by the model are those resulting from direct exhaust emissions, and do not account for either the destruction or photochemical formation of the pollutant in the atmosphere. The model also assumes that the pollutant of interest has emission formation and dispersion characteristics similar to that of CO.

In the SwRI model, the relatively insignificant indoor CO exposures were set to zero. Exposures in three mobile source microenvironments (street canyons, tunnels and parking garages), where elevated concentrations of mobile source pollutants could be experienced, were added to the OAQPS version of the model. Finally, a national extrapolation procedure designed expressly for mobile sources was devised.

There are three required inputs. The first is the nationwide urban and rural populations for the year of interest. The second is a series of 33 emission factors which are described in Table 2-10. The third input is a list of 25 ambient pollutant concentrations, specifying the 24 concentration intervals or bins for which cumulative person-hours of exposure are to be calculated.

The output lists the total annual person-hours of exposure found in each of the specified concentration intervals. Using this information, a mean exposure level may be calculated.

Urban and rural populations for 1986 and 1995 were estimated based on U.S. Department of Commerce data and are given below.[19]

	Urban	Rural	Total
1986	180,000,000	60,000,000	240,000,000
1995	195,000,000	65,000,000	260,000,000

As seen in Table 2-10, the emissions input required for the model are FTP emission factors and an emission factor at an average speed of 10 mph, expressed in grams/minute. For the tunnel microenvironment and rural areas, a 35 mph steady state emission factor will be used. For light-duty vehicles, an idle emission factor is also required. As mentioned previously, these emission factors were chosen to best represent the vehicle operating conditions in each neighborhood/ microenvironment.

The weighted emission factors given in Section 2.3.3 are considered FTP (or transient) emission factors. VMT fractions are then used to determine FTP emission factors for each vehicle class and a composite emission factor for the fleet. The LDV/LDT FTP has an average speed of 19.6 mph; this average speed was used for both LDV/LDT and HDV to convert the g/mile factor to g/min, as required for the model. For LDV/LDT this is certainly correct. For HDV it is less clearly so but is a reasonable approximation.

The VMT fractions in Table 2-8 represent VMT fractions nationwide. Urban and rural VMT fractions differ, particularly for the heavy-duty vehicle classes 7 and 8. For these heavy-duty subclasses, rural VMT is estimated to exceed urban VMT by a factor of 2.7.[15] Since these heavy-duty subclasses are responsible for the majority of diesel particulate emissions, urban and rural VMT fractions will be used. The urban and rural VMT fractions for each vehicle class are given below.[15]

Table 2-10

Model Emission Factor Inputs

Neighborhood or Microenvironment	Assumed Emission Factor (g/min)
Weekday urban residential	FTP
Weekday urban commercial	10 mph
Weekday urban industrial	10 mph
Weekday suburban residential	FTP
Weekday suburban commercial	FTP
Weekday suburban industrial	FTP
Weekday street canyon	10 mph
Weekday tunnel	35 mph
Weekday parking garage	0.5 times idle factor plus 0.5 times 10 mph (LDV only)
Dummy mobile source emission factor	Use tunnel factor as dummy (to allow for future expansion)
Weekday rural	35 mph
Saturday urban residential	0.72* times weekday FTP factor
Saturday urban commercial	0.72 times 10 mph factor
Saturday urban industrial	0.72 times 10 mph factor
Saturday suburban residential	0.72 times FTP factor
Saturday suburban commercial	0.72 times FTP factor
Saturday suburban industrial	0.72 times FTP factor
Saturday street canyon	0.72 times 10 mph factor
Saturday tunnel	35 mph
Saturday parking garage	0.5 times idle plus 0.5 times 10 mph (LDV only)
Dummy mobile source emission factor	Use tunnel factor as dummy
Saturday rural	0.72 times 35 mph factor
Sunday emission factors	In current version of program, Sunday emission factors are equal to Saturday factors.

---

* The factor of 0.72 is used to adjust for relative traffic volume.

	VMT Fractions					
			HDV			
	LDV	LDT	2B	3-5	6	7-8
1986						
Urban	.721	.206	.026	.005	.010	.032
Rural	.617	.247	.016	.003	.008	.109
1995						
Urban	.717	.209	.031	.004	.006	.033
Rural	.613	.250	.018	.002	.005	.112

To determine the 10 mph and 35 mph emissions factors and the idle emission factor for the light-duty diesel vehicle fleet, it was necessary to calculate ratios of emissions at these speeds to FTP emissions. For the light-duty vehicles, data from the New York City Cycle (NYCC), with an average speed of 7.07 mph, were used to represent the 10 mph cyclic emission factor. The results of two studies were used which collectively contained data at idle, steady state speeds of 31 mph, 50 mph, and 53 mph as well as FTP and NYCC data.[20,21] Ratios of emissions at the steady state speeds (g/min) to FTP emissions (g/min) were plotted and found to increase exponentially. The ratio at 35 mph was estimated from the plot.

For heavy-duty vehicles, emissions data from three in-service buses over various chassis dynamometer driving cycles were used to calculate the 35 mph steady state ratio for the tunnel microenvironment and urban areas.[22] Emissions data were taken at idle, steady state speeds of 12.5 mph and 25 mph and over the heavy-duty chassis driving cycle. To obtain the ratio at 35 mph, it was assumed that the ratio from 25 mph to 35 mph would continue to increase linearly. Data were not available at higher speeds to determine whether the ratios would increase exponentially at some point; therefore, the linear assumption may result in a slight underprediction of the 35 mph ratio.

Very little data exist to calculate cyclic emission factors at 10 mph for heavy-duty vehicles. Recent EPA test programs have focused on emissions characterization of in-use transit buses. These programs involved buses which were temporarily removed from operating service and which were tested without additional maintenance in their chassis configurations over test cycles designed specifically to simulate transit bus operation. The test cycles have average speeds ranging from 8.8 to 12.4 mph. Information regarding the test programs can be found in reference 23. An overall average transit bus emission factor was estimated to be 5.52 g/mile, or roughly 0.92 g/min, based on an average speed of 10 mph. A 10 mph/FTP ratio was estimated such that the resulting emission factor would equal 0.92 g/min, based on the 1986 FTP data.

This ratio will be used for both 1986 and 1995 to calculate the heavy-duty contribution in the street canyon neighborhood, where public exposure to transit bus emissions is relatively high.

For the remaining urban industrial and commercial neighborhoods, a 10 mph cyclic emission factor for heavy-duty trucks is required. For these neighborhoods, the composite heavy-duty FTP g/mile emission factor will be used. The ratio at 10 mph is thus equal to the ratio at this speed to the FTP average speed of 19.6 mph. The resulting ratio at 10 mph is 0.51.

The resulting ratios for the light-duty and heavy-duty vehicles are given in Table 2-11. These ratios were applied to the FTP g/min calculated for each vehicle class and then summed to obtain VMT-weighted 10 mph and 35 mph g/min composite emission factors. The emission factor inputs for the 1986 low and high sales scenarios are the same. The reason is that the sales fractions used for the scenarios only differ for the 1985 and 1986 model years. This difference was found to be negligible when computing the model emission factor inputs.

It should be noted that the absolute emission factors and the assumptions used to estimate the off-FTP values directly influence the estimated absolute particulate exposures in 1986 and 1995, but that the percent decrease in particulate exposure from 1986 to 1995 is sensitive only to the more reliable FTP emission factors for the two years.

The final model input required is a list of 25 ambient pollutant concentrations defining the concentration intervals for which cumulative person-hours of exposure are calculated. The chosen concentrations are given in Table 2-11 and range from 0.00 to 50.00 ug/m^3 for 1986 and 0.00 to 31.47 ug/m^3 for 1995.

The modified NEM does not take into account projected increases in VMT. Since most of the modified NEM is based on 1981 monitoring data, projected VMT for 1986 and 1995 will be compared to 1981 VMT and the exposures predicted by the modified NEM adjusted upward accordingly. Since VMT from 1981 to 1986 is expected to increase 10 percent, the 1986 exposures were adjusted upward by 1.10. Similarly, since VMT from 1981 to 1995 is expected to increase 31 percent, the 1995 exposures were adjusted upward by a factor of 1.31.[15] This VMT adjustment is likely to be somewhat conservative on top of the adjustment that is made for population growth, since some of both types of growth will be outward at urban fringes, rather than upward in the areas of current population and VMT concentration. The caveat applies to many of the other pollutants, whose exposure is estimated in the same manner.

Table 2-11

Diesel Particulate Ratios and Model Inputs

	Ratios g/min/g/min Idle/FTP	10 mph/FTP	35 mph/FTP
LDV/LDT	0.33	0.64	0.79
HDV	N/A*	0.51**	0.63

25 Pollutant Concentrations (ug/m^3)

1986

0.00, 0.50, 0.61, 0.75, 0.91, 1.11, 1.36, 1.66, 2.00, 2.50, 3.00, 3.70, 4.50, 5.50, 6.80, 8.20, 10.10, 12.30, 15.00, 18.40, 22.00, 27.00, 33.00, 41.00, 50.00

1995

0.00, 0.13, 0.16, 0.21, 0.26, 0.33, 0.42, 0.54, 0.68, 0.86, 1.10, 1.40, 1.77, 2.25, 2.87, 3.64, 4.63, 5.88, 7.47, 9.50, 12.07, 15.34, 19.49, 24.77, 31.47

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* An idle emission factor is used to simulate emissions in a parking garage. It is assumed there are no HDV in parking garages; therefore, a HDV idle/FTP ratio is not applicable.

** For the street canyon microenvironment, a 10 mph/FTP ratio of 2.50 is used. For 1986, this ratio results in an emission factor of 0.92 g/min, equal to a current estimated transit bus emission factor. The same ratio is applied in 1995.

The mean exposure levels predicted by the model, adjusted to take into account projected increases in VMT, are given below.

Exposure (ug/m^3)

	Urban	Rural	Nationwide
1986	2.63	2.38	2.56
1995 Low Sales	1.27	1.06	1.22
1995 High Sales	1.69	1.27	1.58

As seen above, the rural exposures are quite similar to the urban exposures. This is due to the greater fraction of heavy-duty vehicle classes 7-8 in rural areas, resulting in a higher composite emission factor. In addition, the modified NEM calculates rural exposures quite crudely. This is a weakness of the model and should be taken into consideration when reviewing the results.

2.4.2 Comparison to Other Exposure Estimates

This section compares the mean exposure levels predicted by the modified NEM to three other diesel particulate exposure estimates made previously. These are: 1) diesel particulate exposures estimated by OMS in a somewhat similar manner in 1983, 2) an estimate made by the Lovelace Inhalation Toxicology Research Institute (ITRI) in 1981, and 3) exposures estimated by OMS in 1983 using a lead surrogate approach. The comparison among these methods also holds for other pollutants in this report for which the modified NEM was used.

2.4.2.1 1983 OMS Exposure Estimate

In 1983, OMS projected diesel particulate exposure in urban areas for 1995.[8] OMS used the original CO NEM, with the only modification being the removal of all indoor sources. The annual average CO exposure was 2.12 ppm. An average annual exposure for diesel particulate for 1995 was then estimated by ratioing CO and diesel particulate emission factors and multiplying the result by 2.12 ppm. The CO emission factor was 62.3 g/mile. This is the CO national average emission factor for 1978, which is the same year as the CO NEM data base. The composite diesel particulate emission factor used (for the best estimate base sales scenario) was 0.0554 g/mile. Since VMT from 1978 to 1995 was expected to increase 45 percent, the diesel particulate emission factor was adjusted upward by 1.45.

The projected exposure in 1995 for the best estimate base sales scenario is 1.5-1.6 ug/m^3 from LDDV and 1.6-2.1 ug/m^3 for HDDV. The total projected exposure is 3.1-3.7 ug/m^3. This is considerably higher than the urban exposure range of 1.3-1.7 ug/m^3 estimated in this study. The urban exposure of 1.7 ug/m^3 estimated in this study corresponds to an FTP composite diesel particulate emission factor of 0.053 g/mile. This is quite similar to the composite emission factor calculated in 1983. It appears that the modified NEM calculates lower exposures per g/mile that the CO NEM used previously. The modified NEM does include more recent monitoring data, including an expanded number of monitors, relative to the CO NEM, which may account for the difference.

2.4.2.2 1981 Lovelace Inhalation Toxicology Research Institute (ITRI) Exposure Estimate

ITRI's exposure estimate was confined to LDDV. ITRI used a particle dispersion model to project future atmospheric concentrations of LDDV particulate in urban and rural areas of the U.S. in 1995.[24,25] The atmosphere is represented by a grid of cells with variable heights that move with the velocity and direction of wind. Atmospheric concentrations of particles were calculated by sweeping the grid of cells, beginning on the upwind side of the urban area and passing them across the city. The model accounts for emissions as well as turbulent mixing, particle diffusion, particle settling and particle resuspension. Assumptions include: 1) the population was distributed with the same density as diesel vehicles, 2) the typical U.S. city has 40 percent of the population in the city center during work hours and has an average wind speed of 5 m/sec, and 3) particles entering a cell are assumed to be mixed uniformly throughout the cell.

Information used included the current (1981) land areas, populations and gasoline consumption rates for all major U.S. cities and standard metropolitan statistical areas. These data were scaled to represent conditions after 1995 when 20 percent of all LDV were assumed to be diesel-powered. ITRI projected average concentrations of LDDV particulate based on assumed LDDV emission rates of 0.2 g/mile and 0.5 g/mile. This comparison will be limited to the results obtained with the 0.2 g/mile emission rate, since this emission rate agrees with that used in this study and the previous OMS study.

Average concentrations of LDDV particulate in urban and rural areas were projected to be 0.2 ug/m^3 and 0.02 ug/m^3, respectively, based on an emission rate of 0.2 g/mile. In addition to projecting these "background" atmospheric concentrations in urban and rural areas, calculations were also made of the higher concentrations that may occur near urban street canyons and expressways. The exposure estimates developed by ITRI are given below:

	Particulate Exposure (ug/m^3) Based on 0.2 g/mile
Typical urban resident	0.2
Urban residents near freeway	2.0
Workers on urban freeway	15.0
Workers in urban street canyon	15.0

The exposures in urban areas and near street canyons and expressways were weighted, with the bulk of the resulting total exposure being due to the urban background, and used in ITRI's risk assessment. The composite LDDV urban particulate exposure was not given by ITRI but has been calculated by EPA to be roughly 0.50 ug/m^3.

The exposure estimate projected by ITRI is lower than the estimates projected in this study; however, the different assumptions used in both studies make a direct comparison difficult. ITRI assumed 20 percent of all LDV were diesels. In comparison, this study assumed 0.9-11.5 percent of LDV sales were diesels. ITRI therefore assumed a much greater penetration of LDDV. Heavy-duty vehicles were not considered by ITRI. In this study, heavy-duty vehicles are responsible for 66-90 percent of the 1995 urban exposure depending on diesel sales. (These percentages were derived using the g/mile emission factors in Section 2.3.3 together with the 1995 urban VMT fractions in Section 2.4.1.)

2.4.2.3 Exposure Estimate Based on a Lead Surrogate Approach

For comparison, in 1983, EPA also used 1975 atmospheric lead monitoring data as a surrogate to estimate atmospheric levels of diesel particulate in 1995.[8] Estimates were provided of ambient diesel particulate concentrations at one or two particular monitor locations in a large number of U.S. cities. The monitors were chosen in areas having no large stationary sources of lead.

For this analysis, an estimate was made of the fleet's automotive lead emission factor which caused the observed ambient lead levels, and is compared to the expected diesel particulate emission factor. The 1995 projected ambient diesel particulate concentration in 1995 was set equal to the urban ambient lead concentration in 1975 multiplied by the ratios of 1995 diesel particulate to 1975 lead emission factors, dispersion factors and VMT. Even though the 1995 fleet average (best estimate sales) diesel particulate emission factor is less than half the 1975 fleet average lead emission factor, ambient concentrations of diesel particulate in 1995 were projected to be 1.63 times the urban ambient lead concentrations in 1975. This is due to the projected increase in VMT (60 percent) in 1995 and the higher estimated dispersion factor for diesel particulate relative to lead (1.0 versus 0.43). Resulting ambient diesel particulate concentrations varied from 0.7 to 3.9 ug/m^3, depending on the city and particular monitor. The estimates projected in this study, as well as the earlier OMS estimates, fall within this range.

2.4.3 Comparison to Particulate Monitoring Data

The annual geometric mean total suspended particulate (TSP) concentration in 1984 was 50 ug/m^3.[17] It has remained fairly constant at this level since 1982. The nationwide average diesel particulate concentration in 1986 calculated above is roughly 5.1 percent of the 1984 total. This percentage is similar to that estimated for emissions (3.9 percent) in Section 2.3.5.

PM_{10} denotes particulate matter less than 10 microns in size. Since virtually all diesel particulate falls under PM_{10}, it would be useful to estimate the contribution of diesel particulate to PM_{10} concentrations. A PM_{10}/TSP ratio of 0.50 is used, based on an examination of 1982-1983 monitoring data.[26] Applying this ratio to the 1984 TSP concentration results in an annual mean PM_{10} concentration of 25 ug/m^3. Thus, diesel particulate emissions appear to account for roughly 11 percent of the annual mean PM_{10} concentrations. It should be pointed out, however, that this comparison is probably misleading since the TSP and PM_{10} data are all at fixed site monitors. At these sites, the diesel levels are probably much higher than the mean personal estimates given in this report.

2.5 Health Effects of Diesel Particulate and Unit Risk Estimates

The mutagenic activity of organic extracts from diesel particulate was first reported in 1979. At that time, the limited data available from animal and epidemiology studies were not sufficient for a cancer risk assessment. As a result, EPA conducted a large research program to evaluate the health effects associated with exposure to diesel emissions.

A comparative potency method was used to determine a unit risk estimate for diesel particulate. In this method, the potency of the organic extracts from diesel particulate are compared with the potencies of extracts from sources for which epidemiological data are available. A large number of mutagenesis and carcinogenesis studies were performed to determine relative potencies. The methodology and results are discussed in detail in reference 7. The unit risks for the light-duty diesel particulate sources ranged from

2.0×10^{-5}–3.5×10^{-5}. These represent the means of the lower and upper 95 percent confidence limits. The upper confidence limit unit risks ranged from 3.3×10^{-5}–6.3×10^{-5}.

Two other analyses of EPA's comparative potency data were performed by Dr. Harris for NAS [27] and by the Lovelace Inhalation Toxicology Research Institute (ITRI).[24] Harris' mean estimate was a 0.0035 percent proportional increase in risk per year per ug/m^3 exposure assuming lifetime exposure. Harris' relative risk was translated into an absolute measure of lung cancer incidence using the methodology described in reference 28. The result is a unit risk of 1×10^{-4}. Harris' upper confidence limit estimate (0.0252 percent proportional increase in risk) is roughly equivalent to a unit risk of 7.5×10^{-4}.

ITRI calculated a range of unit risks which differ depending on which comparative source was used. The resulting range was 4.9×10^{-6} to 2.1×10^{-4}. These risks appear to be upper confidence limits. ITRI chose 7.0×10^{-5} as being most representative.

The range of unit risks that will be used in this paper is 2.0×10^{-5} to 1.0×10^{-4}. Assuming an average lifetime of 70 years, the range of risk estimates, on an annual basis, is 0.29×10^{-6} to 1.4×10^{-6} lung cancers per person per ug/m^3 particulate. The lower end of the range is the lowest EPA estimate. The upper end of the range is Harris' mean estimate which is also in rough agreement with the ITRI estimate. The range incorporates both EPA's and ITRI's upper confidence limit estimates. Harris' upper confidence limit estimate was not included due to the uncertainty of translating his relative risk into an absolute measure of risk.

The comparative potency studies that are the basis of the risk estimates used the organics extracted from diesel particulate. Inhalation studies using whole diesel emissions (i.e., particulate and gas phase emissions) were also performed concurrently but the results were negative or inconclusive.[29]

Long-term animal inhalation studies are presently being conducted by ITRI, Fraunhofer, Battelle-Geneva and Battelle-Northwest. In contrast to the previous studies, preliminary results indicate that lung tumors have been found at concentrations no higher than those tested with negative results in the previous studies. Reports are expected shortly.

2.6 Current and Projected Health Risk

The annual risk estimates are combined with the exposure estimates (given in Section 2.4.1) and population estimates to obtain estimates of lung cancer incidence for 1986 and 1995. The results are given in Table 2-12.

As seen in this table, the total risk in 1986 ranges from 178-860 cancer incidences and drops from 33-48 percent in 1995, depending on projected diesel sales.

Table 2-12

Annual Lung Cancer Risk from Diesel Particulate Exposure

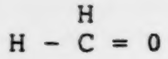
		Urban	Rural	Total
1986		137-661	41-199	178-860
1995	Low Sales	72-346	20- 97	92-443
1995	High Sales	95-461	24-115	119-576

3.0 FORMALDEHYDE

3.1 Formation, Composition and Control Technology

Formaldehyde is the most prevalent aldehyde in vehicle exhaust and is formed as a result of incomplete combustion of the fuel. Formaldehyde is emitted in the exhaust of both gasoline and diesel-fueled vehicles. It is not a component of evaporative emissions. Formaldehyde is of interest due to its photochemical reactivity and suspected carcinogenicity.

Formaldehyde has the chemical formula HCHO. Its structure is shown below.



$$H - \overset{H}{C} = 0$$

Use of a catalyst has been found to be effective for controlling formaldehyde emissions. Formaldehyde emissions are controlled to roughly the same extent as total hydrocarbon emissions with a catalyst.

3.2 Emissions

In this section, formaldehyde emission factors (g/mile) for calendar years 1986 and 1995 are presented. Nationwide mobile source formaldehyde emissions (metric tons/year) for 1986 and 1995 are then calculated by combining the emission factors with VMT data for 1986 and 1995. The results are then compared to estimates of nationwide total formaldehyde emissions.

3.2.1. Emission Factors for Calendar Years 1986 and 1995

For this analysis, formaldehyde emissions for the various classes are expressed as a percentage of total hydrocarbons. These percentages were then applied to MOBILE3 exhaust hydrocarbon (HC) output for 1986 and 1995 to obtain the formaldehyde emission factors. This was done for two reasons: 1) formaldehyde emissions can vary considerably within a vehicle class but are more consistent when expressed as a percentage of total hydrocarbons, and 2) use of the MOBILE3 HC output should more accurately represent in-use emissions since deterioration and effects of malfunction and tampering/misfueling are accounted for in MOBILE3.

An important issue to address is whether the percentages chosen are adequate to use for the excess hydrocarbons that come from deterioration, malfunction and tampering/misfueling. Effects of deterioration, malfunction and tampering/misfueling have been studied to the greatest extent with LDGV; therefore, LDGV data are examined here.

The percentages for LDGV were based on data for both new vehicles and in-use low and high mileage vehicles. The percentages do, therefore, factor in the effects of deterioration and minor malfunctions. The percentages are remarkably similar for non-catalyst-equipped vehicles as well as vehicles equipped with oxidation catalysts and 3-way plus oxidation catalysts. Overall, formaldehyde emissions varied from roughly 1-3 percent of total HC while HC emissions varied from roughly 0.2 -6.0 g/mile. Even within a particular vehicle category, wide variations in HC emissions generally have little effect on the formaldehyde percentages. For example, HC emissions from two in-use oxidation catalyst-equipped vehicles varied from 0.4 g/mile to 4.7 g/mile, yet the formaldehyde percentage for both vehicles was 1.5 percent.[30] It is true that the formaldehyde percentage itself is subject to some variation, but the variation is slight compared to the variations in the absolute level of formaldehyde emissions.

Studies have specifically examined the effects of misfueling and malfunctions on formaldehyde and other exhaust emissions. In a misfueling study, exhaust emissions from a catalyst-equipped vehicle were measured initially with unleaded fuel.[31] Total aliphatic aldehydes were measured but can be used as an indicator of formaldehyde emissions. The vehicle was then driven 5000 miles on various commercial leaded gasolines. Total aliphatic aldehyde emissions initially (with unleaded fuel) were 2.1 percent of the total HC emissions. Following 5000 miles of misfueling, the percentage increased only slightly to 2.5 percent.

Malfunctions that have been evaluated include, but are not limited to, 12 percent misfire, disabled EGR, rich best idle and high oil consumption. Results with non-catalyst-equipped vehicles indicate roughly similar formaldehyde percentages with and without malfunctions.[32] With catalyst-equipped vehicles, formaldehyde emissions tend to be reduced with malfunctions while total hydrocarbon emissions increase, resulting in lower formaldehyde percentages with malfunctions.[33-35]

In summary, formaldehyde percentages appear relatively stable over a wide range of operating conditions and HC emissions. The data support the use of expressing formaldehyde emissions as a percentage of total hydrocarbons.

Formaldehyde emissions, expressed as a percentage of total hydrocarbons, for the vehicle classes in 1986 and 1995 are given below:

Vehicle Class	% of Total Exhaust HC	
	1986	1995
LDGV	1.3	1.0
LDGT 1,2	1.3	1.1
LDDV	4.1	4.1
LDDT	4.1	4.1
HDGV	3.1	3.1
HDDV-TRUCK	3.0	3.0
HDDV-COMM. BUS	10.0	10.0

Data for the light-duty gasoline-fueled vehicles were taken from references 30 and 32-36. These data were assumed to apply to light-duty gasoline-fueled trucks as well. These references include data for both new and in-use vehicles. The decreasing percentages for these vehicles from 1986 to 1995 reflect the phase-out of non-catalyst-equipped vehicles. Since a greater percentage of non-catalyst-equipped trucks than vehicles are projected in 1995, the percentages for trucks do not decrease as much in 1995 as the vehicles.

Data for the light-duty diesel vehicles were taken from references 37 and 38. Again, these data were assumed to apply to light-duty diesel trucks as well. The formaldehyde percentage of total HC is assumed to remain constant from 1986 to 1995.

Data for the heavy-duty gasoline- and diesel-fueled engines were taken from references 22 and 38. Since the heavy-duty diesel bus engines appeared to have greater percentages than the other heavy-duty diesel engines, they were treated separately, and assigned a VMT fraction of 0.002.[17]

Applying these percentages to the MOBILE3 exhaust HC emission factors by vehicle class for 1986 and 1995 and using MOBILE3 VMT fractions, resulting composite FTP formaldehyde emission factors are given below.

	FTP g/mile	
	1986	1995
with I/M	0.0418	0.0201
without I/M	0.0453	0.0224

Note that MOBILE3 runs were used which assumed both the presence and absence of an Inspection/Maintenance (I/M) program. An I/M program should have a beneficial impact on formaldehyde emissions since formaldehyde emissions are correlated with HC emissions. The I/M program selected has the following characteristics:

Start year (January 1): 1983
Pre 1981 MYR stringency rate: 20%
Mechanic training program?: No
First MYR covered: 1951
Last MYR covered: 2020
Vehicle types covered: LDGV
1981 and later MYR test type: Idle
1981 and later MYR test cutpoints: 1.2% ICO/220 ppm IHC

This represents the minimum EPA requires of I/M programs.

3.2.2 Nationwide Mobile Source Formaldehyde Emissions

Nationwide mobile source formaldehyde emissions (metric tons/year) for 1986 and 1995 are calculated by combining the calendar year emission factors in the previous section with projected VMT data for 1986 and 1995. The VMT data are given in Table 2-8.

The nationwide mobile source formaldehyde emissions for 1986 and 1995 are given below.

Mobile Source Formaldehyde Emissions
(metric tons per year)

	1986	1995
with I/M	68,003	38,880
without I/M	73,697	43,329

As seen, formaldehyde emissions decrease from 1986 to 1995 due to the projected HC reductions.

It should be noted that these estimates only account for formaldehyde directly emitted. Formaldehyde formed indirectly from photooxidation of mobile source volatile organic compounds (VOC) is not included but will be addressed in a later section.

3.2.3 Other Sources of Formaldehyde

Formaldehyde is produced in the U.S. by 14 chemical companies in 48 locations encompassing 21 states. Formaldehyde is used in the manufacture of four major types of resins: urea-formaldehyde, melamine-formaldehyde, phenol-formaldehyde and polyacetal resins. These resins are used in a wide variety of products, such as plywood, particle board and counter tops. Formaldehyde is also used as a raw material in several synthetic organic chemical production processes.

In addition, formaldehyde is produced as a by-product in the following types of processes: combustion (mobile, stationary and natural sources), petroleum refinery catalytic cracking and coking, phthalic anhydride production and atmospheric photooxidation of unburned hydrocarbons. Table 3-1 provides a summary of 1985 estimated formaldehyde emissions by source category.[39] The contribution of atmospheric photooxidation is extremely uncertain and therefore not included in Table 3-1.

The estimates of mobile source formaldehyde emissions in 1986 with and without I/M were 68,003 and 73,697 metric tons/year, respectively. This represents 26-29 percent of the total formaldehyde emissions given in Table 3-1. Therefore, roughly 26-29 percent of the formaldehyde emissions emitted directly into the air (versus formed photochemically) appear to be attributable to mobile sources.

3.3 Ambient Concentrations of Formaldehyde Emitted by Mobile Sources

In this section, urban and rural concentrations of formaldehyde emitted from vehicle exhaust are estimated for 1986 and 1995, using the modified version of the CO NEM discussed previously. The contribution of mobile sources to ambient formaldehyde levels is then discussed.

3.3.1 Estimation of Urban and Rural Exposure

The three required model inputs are: 1) the nationwide urban and rural populations for the year(s) of interest, 2) FTP, 10 mph, 35 mph, and LDGV idle emission factors (g/min), and 3) a list of 25 ambient pollutant concentrations, defining the concentration intervals for which cumulative person-hours of exposure are to be calculated. The first input is given in Section 2.4.1. Similarly, the FTP emission factors have also already been provided and are simply converted from g/mile to g/min using the average FTP speed of 19.6 mph. To calculate emission factors at 10 mph and 35 mph, MOBILE3 runs were made at these speeds and the ratios of the total exhaust HC emissions at these speeds to those at the FTP average speed were determined. These speed correction factors were then multiplied by the composite FTP emission factors to obtain the emission factors at 10 mph and 35 mph. The speed correction factors were 1.75 for 10 mph and 0.54 for 35 mph.

The idle LDGV emission factors for 1986 and 1995 are 0.0009 and 0.0006 g/min, respectively. Idle emission factors of both HC and formaldehyde have been measured less often than those for the FTP, and the formaldehyde:HC ratio approach was not used for idle. They were instead determined directly using the formaldehyde emission rate data in reference 40 together with projected LDGV VMT fractions of non-catalyst-equipped and catalyst-equipped vehicles in 1986 and 1995. In 1986, 12

Table 3-1

Summary of Estimated 1985 Formaldehyde Emissions by Source Category

Source	Formaldehyde Emissions (metric tons/year)
Direct Producers	910
Resin Manufacture	2,789
Synthetic Chemical Production	655
Indirect Producers*	
Combustion (Mobile, Stationary, Natural)	250,000
Petroleum Catalytic Cracking	3,200
Phthalic Anhydride Production	1
TOTAL:	257,555

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* Excludes atmospheric photooxidation due to the uncertainties inherent in the estimate.

percent of the LDGV VMT is projected to be due to non-catalyst-equipped vehicles; in 1995, the percentage drops to 0.2 percent.

The 25 ambient pollutant concentrations for 1986 ranged from 0.0 to 24.0 ug/m^3. For 1995, they ranged from 0.0 to 10.0 ug/m^3. Logarithmic intervals were chosen to maximize resolution over the entire range.

The mean exposure levels predicted by the model, adjusted to account for increased VMT, are given below. These exposure levels account for direct emissions from mobile sources only.

	Formaldehyde Exposure (ug/m^3)		
	Urban	Rural	Nationwide
1986			
With I/M	1.21	0.56	1.04
Without I/M	1.30	0.60	1.13
1995			
With I/M	0.68	0.31	0.59
Without I/M	0.76	0.35	0.65

It should be noted that these model predictions were made assuming that formaldehyde has emission formation and dispersion characteristics similar to that of CO. The model also does not account for photochemical reactions. The exposure levels predicted by the model are those resulting from direct exhaust emissions, and do not account for either the destruction or photochemical formation of formaldehyde in the atmosphere.

3.3.2 Contribution of Mobile Sources to Ambient Formaldehyde Levels

Ambient formaldehyde levels are a result of formaldehyde directly emitted by sources and formaldehyde formed from photooxidation of VOC. The mobile source contribution to ambient formaldehyde levels also contains both components. The previous sections have attempted to quantitate the directly emitted component. It appears that roughly 26-29 percent of directly emitted formaldehyde may be attributable to mobile sources.

Formaldehyde formed photochemically is much more difficult to quantify. One approach being used by EPA is to determine the relative contribution of various sources based on estimates of annual U.S. VOC emissions for each source. These estimates are given in Table 3-2.[41] Mobile sources account for 30 percent of the total VOC emissions. If it is assumed that the VOC from all sources have the equivalent potential to form formaldehyde, then 30 percent of the formaldehyde formed photochemically is due to mobile sources. It should be noted,

Table 3-2

Sources of VOC Emissions and 1985 National VOC Air Emission Estimates*

Source	VOC Emissions (metric tons/year)	% of Total
Mobile Sources	7,200,000	30
Misc. Solvent Uses	3,600,000	15
Hazardous Waste Treatment, Storage and Disposal Facilities	3,500,000	14
Surface Coating	3,160,000	13
Petroleum Marketing	2,230,000	9
Petroleum Refining	740,000	3
Chemical Manufacture	500,000	2
Industrial Processes	365,000	2
Miscellaneous Sources	3,020,000	12
TOTAL:	24,315,000	

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* Reference 41.

however, that mobile sources are estimated to account for a higher percentage of the total VOC in urban areas. Examination of 1983 VOC emissions in 61 urban ozone non-attainment areas showed the average mobile source VOC contribution to fall between 40-50 percent. [42]

Since the mobile source contribution to both directly emitted and photochemically produced formaldehyde nationwide is roughly equivalent, as a rough approximation, 30 percent of the ambient formaldehyde concentration could be due to mobile sources on a national basis. The mobile source contribution will vary depending on vehicle use in a particular area, season and a variety of other meteorological factors. In the winter, for example, formaldehyde emissions from mobile sources are expected to increase whereas photochemical reactions are expected to be minimized. The mobile source contribution to ambient formaldehyde emissions in the winter could, therefore, be greater.

Photochemical modeling is one approach to use in attempting to determine the relative contributions of mobile and stationary sources to ambient formaldehyde concentrations. Photochemical modeling has been done to simulate meteorology and photochemistry occurring during summer months due to the interest in ozone. As part of one modeling study, designed to determine the ozone impact of methanol-fueled vehicle substitution in California's South Coast Air Basin (SCAB) during a severe ozone episode, mobile source VOC emissions were removed and the resulting formaldehyde concentrations determined.[43] The result indicated that 23 percent of the ambient formaldehyde concentration is due to direct formaldehyde emissions from mobile sources and formaldehyde formed photochemically from mobile source VOC. This result lends some support to the previous estimate. Mobile source VOC in the SCAB is roughly 50 percent of the total which is somewhat higher than most areas of the country. Thirty percent, therefore, can be considered an upper estimate.

3.4 Health Effects of Formaldehyde and Unit Risk Estimates

Formaldehyde can cause a number of acute adverse health effects such as eye, nose, throat and skin irritation, headaches and nausea, as well as death. Formaldehyde has also been found to cause nasal cancer by inhalation in males and females of one strain of rat and in males of another strain, and there is evidence of its carcinogenicity in mice. Human data are more limited. EPA has classified formaldehyde as a probable (B1) carcinogen in humans.

The unit risk estimates derived by EPA are based on a single animal inhalation study conducted by the Chemical Industry Institute of Toxicology (CIIT).[44] In this study, statistically significant increased levels of squamous cell carcinomas (malignant tumors) were found in the nasal cavities of rats at 14.3 ppm. (This is roughly 1000 times ambient levels.) The smaller increase in these carcinomas observed in mice was not statistically significant. In addition to the squamous cell carcinomas, small numbers of benign tumors characterized as polypoid adenomas were observed in rats at each dose level.

The upper confidence limit unit risk estimates based on these data are 1.7×10^{-4} for benign tumors and 1.3×10^{-5} for malignant tumors.[45] The unit risk for malignant tumors of $1.3x10^{-5}$ was used for this analysis. The current consensus in EPA favors use of malignant data only. EPA's Office of Toxic Substances is using the unit risk based on malignant tumors in assessing the risk to garment workers and home residents.[45] Assuming an average lifetime of 70 years, the annual risk estimate is $1.9x10^{-6}$.

3.5 Current and Projected Health Risk

The annual risk estimate is combined with the exposure estimates (given in Section 3.3.1) and population estimates to obtain estimates of cancer incidence for 1986 and 1995. The results are given in Table 3-3.

As seen in this table, the total risk in 1986 ranges from 46-50 cancer incidences and drops to 29-31 cancer incidences in 1995. This decrease is accounted for by the anticipated decrease in HC emissions.

These risk estimates are for formaldehyde emitted directly from vehicle exhaust. Unfortunately, the model used to determine ambient concentrations could not account for the destruction and photochemical formation of formaldehyde in the atmosphere.

An approach to include photochemistry is to use actual ambient monitoring data and assign a mobile source fraction. This approach accounts for formaldehyde directly emitted and formed or destroyed photochemically. OAQPS calculated an urban population weighted average of 12.71 ug/m^3 based on data from four cities.[46] It represents the average of the population weighted summer average in Baltimore, Los Angeles and New Jersey with the winter average in Philadelphia. The summer concentrations used represent maximum rather than average values. As a result, 12.71 ug/m^3 is closer to an urban population weighted maximum concentration. It may also be biased high since these cities are in ozone non-attainment areas. It will be used here to represent a plausible upper limit. A concentration of 1.50 ug/m^3 was selected to represent rural areas.[46]

Table 3-3

Annual Cancer Risk From Direct Emissions of Formaldehyde from Mobile Sources

	Urban	Rural	Total
1986			
With I/M	40	6	46
Without I/M	43	7	50
1995			
With I/M	25	4	29
Without I/M	27	4	31

In Section 3.3.2, it was indicated that mobile sources constituted roughly 30 percent of formaldehyde emitted directly as well as 30 percent of the total VOC emitted. Assuming the VOC from all sources have the equivalent potential to form formaldehyde, a mobile source fraction of 0.30 is selected and applied to the estimated urban and rural concentrations above. Resulting urban and rural formaldehyde concentrations due to mobile sources are 3.81 and 0.45 ug/m^3, respectively.

In comparison, the urban and rural formaldehyde concentrations obtained from modeling are 1.21-1.30 and 0.56-0.60 ug/m^3, respectively. The urban concentration estimated from ambient monitoring data is roughly three times greater than that obtained by modeling. Two possible reasons are: 1) the ambient monitoring data accounts for formaldehyde formed photochemically, and 2) the ambient data may be from fixed site monitors that overrepresent 24-hour exposures of the population. In contrast, the rural concentration obtained from modeling is slightly higher than that estimated from ambient monitoring data. The rural concentrations were estimated rather crudely with both approaches.

The exposure estimates based on ambient monitoring data are then combined with the annual risk estimates and urban and rural population estimates to obtain estimates of lung cancer incidence from current mobile sources. The results are given below:

Annual Lung Cancer Risk
(accounts for photochemical reactions)

	Urban	Rural	Total
Current Mobile Sources	126	5	131

Mobile sources currently could be responsible for as many as 131 cancer deaths from formaldehyde exposure. Direct emissions of formaldehyde were projected to decrease roughly 42 percent from 1986 to 1995. Mobile source VOC follows a similar trend. If it is assumed that the photochemical component is also reduced similarly, the mobile source risk in 1995 is estimated be roughly 77 cancer deaths.

3.6 Current Activities

Research activity is planned or underway in three areas: 1) emissions characterization, 2) photochemistry, and 3) ambient monitoring. Formaldehyde emissions from mobile sources are being characterized under cold temperature conditions, since it appears formaldehyde emissions could increase under these conditions. In addition, work is planned in FY87 to investigate factors affecting formaldehyde formation in the atmosphere. Specifically, smog chamber data will be analyzed to rank order the various organic species based on their ability to generate formaldehyde. Modeling simulations will be conducted to assess the effect of varying light intensity, temperature and other meteorological parameters on formaldehyde yields. Smog chamber experiments will also be conducted to determine how well two major chemical mechanisms predict formaldehyde formation.

In the monitoring area, formaldehyde measurements will be included in EPA's Toxic Air Monitoring System (TAMS) network. Measurements will initially be made in three cities (Houston, Boston and Chicago) with a single monitoring site in each city. TAMS should eventually be operational in three to five cities with up to three to four monitors in each city.

It should be noted that, in the case of formaldehyde, formaldehyde levels inside many homes are significantly greater than outside levels. Recent monitoring results indicate that formaldehyde levels in new (less than one year old) conventional homes generally fall within the range of 62 ug/m^3 to 250 ug/m^3; few measurements exceeded 375 ug/m^3. In new mobile homes, formaldehyde levels generally fall within the range of 250 ug/m^3 to 375 ug/m^3 with the highest levels measured near 500 ug/m^3.[45] EPA has calculated expected 10-year averages for formaldehyde levels in homes built today, acknowledging that there is a significant source of uncertainty associated with the estimates. The calculated 10-year averages are 88 ug/m^3 for conventional homes built using significant amounts of ureaformaldehyde pressed wood and 125 ug/m^3 for mobile homes.[45] These exposures are significantly higher than current outdoor urban exposures.

4.0 BENZENE

The Office of Mobile Sources has recently completed a thorough analysis on the carcinogenic impact of benzene emissions.[47] This section will summarize that analysis. Also discussed is an alternative approach to estimating the risk using ambient monitoring data in conjunction with emissions data. A risk analysis performed by the California Air Resources Board (CARB) is then summarized and compared with the risk estimates presented in this report.

4.1 Formation, Composition, and Control Technology

Benzene is an aromatic hydrocarbon with the formula C_6H_6. It is present in both exhaust and evaporative emissions. Current data show the benzene level of current gasoline to be about 1.3%, with diesel fuel containing insignificant levels of benzene. Very little exhaust benzene is unburned fuel benzene. Some work indicates that non-benzene aromatics in the fuels cause about 70-80% of the exhaust benzene formed. Benzene also forms from engine combustion of non-aromatic fuel hydrocarbons. The fraction of benzene in the exhaust varies depending on control technology and fuel composition but is generally about 3-5%. The fraction of benzene in the evaporative emissions also depends on control technology (e.g., whether the vehicle has fuel injection or a carburetor) and fuel composition (e.g., benzene level and RVP) and is generally about 1%. These data also show that diesel vehicles account for only about 3% of the total mobile source benzene emitted.

4.2 Emissions

4.2.1 Emission Factors for Calendar Years 1985 and 1995

An approach similar to that described earlier for formaldehyde was employed for benzene. Benzene emissions were expressed as a percentage of exhaust and evaporative hydrocarbons. These percentages were then applied to the hydrocarbon emissions data in MOBILE3 to obtain composite emission factors for calendar years 1985 and 1995. For the purpose of this report, it will be assumed that calendar year emission factors for 1985 and 1986 will not differ significantly.

Benzene emissions, expressed as a percentage of exhaust and evaporative emissions for the various vehicle classes, are given in Table 4-1.

The test vehicles used to determine the benzene percentages were low mileage, well maintained vehicles. Like formaldehyde, it is important to address whether the percentages chosen are adequate to use for the excess hydrocarbons that come from deterioration, malfunction and tampering/misfueling.

Table 4-1

Benzene Emissions Expressed as Percentages of HC

Vehicle Class	% of Exhaust HC	% of Evap. HC
LDGV		0.35-1.53*
3 Way Cat	5.12	
3 Way + Ox Cat	2.78	
Non-Cat or Ox Cat	3.95	
LDGT1,2	3.24	1.1
LDDV	2.40	
LDDT	2.40	
HDGV	3.48	1.1
HDDV	1.10	

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* 1.11-1.53% for carbureted LDGV.
0.35-0.46% for fuel injected vehicles.

For non-malfunction conditions, there did not appear to be any consistent differences in benzene percentages among the various control technologies (non-catalyst, oxidation catalyst, 3-way catalyst, and 3-way plus oxidation catalyst), regardless of whether the vehicles were tested new or in-use. There were some differences in benzene percentages found between certain malfunction modes and the unmodified test conditions. The most significant of these were 1) the 12 percent misfire mode which consistently decreased the benzene percentage (while greatly increasing total HC) and 2) the rich best idle mode which consistently increased the benzene percentage as well as the total HC (but not as much as the total HC increase in the 12% misfire mode). Due to the lack of differences among control technologies, the offsetting nature of these two malfunctions, and the lesser effects of the other malfunctions, no adjustments were made to the benzene percentages for malmaintenance/tampering.

Resulting composite FTP benzene emissions for 1985 and 1995 are 0.128-0.135 g/mile and 0.088-0.094 g/mile, respectively. The ranges result from the ranges given for the percentage of evaporative hydrocarbons for LDGV. The MOBILE3 runs assumed the presence of a standard, minimum I/M program. As stated previously, diesel vehicles account for a small percentage (3%) of the total mobile source benzene emissions.

Based on the OMS analysis, RVP control, which would be accompanied by a small increase in both benzene content and total aromatic content of gasoline, would have little or no effect on overall fleet emissions or on the number of cancer incidences.[47]

4.2.2 Contribution of Mobile Sources to Nationwide Benzene Emissions

In 1982, total benzene emissions were roughly 294,000 metric tons.[47] Mobile sources account for 250,000 metric tons, or 85 percent of the total. Of the mobile source contribution, 70% comes from exhaust of motor vehicles while 14% of the benzene emissions are motor vehicle evaporative emissions. About 1% of the total benzene emissions occur during motor vehicle refueling. The remaining benzene emissions come from stationary sources with coke ovens being responsible for 10% of the total. Obviously, in regions without coke ovens virtually all benzene is from mobile sources.

4.3 Health Effects of Benzene and the Unit Risk Estimate

Several epidemiology studies on workers exposed to benzene have identified benzene as a carcinogen causing leukemia in humans. The upper confidence limit unit risk estimate has been determined from these studies to be 8.5×10^{-6}.[47,48]

For non-malfunction conditions, there did not appear to be any consistent differences in benzene percentages among the various control technologies (non-catalyst, oxidation catalyst, 3-way catalyst, and 3-way plus oxidation catalyst), regardless of whether the vehicles were tested new or in-use. There were some differences in benzene percentages found between certain malfunction modes and the unmodified test conditions. The most significant of these were 1) the 12 percent misfire mode which consistently decreased the benzene percentage (while greatly increasing total HC) and 2) the rich best idle mode which consistently increased the benzene percentage as well as the total HC (but not as much as the total HC increase in the 12% misfire mode). Due to the lack of differences among control technologies, the offsetting nature of these two malfunctions, and the lesser effects of the other malfunctions, no adjustments were made to the benzene percentages for malmaintenance/tampering.

Resulting composite FTP benzene emissions for 1985 and 1995 are 0.128-0.135 g/mile and 0.055-0.057 g/mile, respectively. The ranges result from the ranges given for the percentage of evaporative hydrocarbons for LDGV. The MOBILE3 runs assumed the presence of a standard, minimum I/M program. As stated previously, diesel vehicles account for a small percentage (3%) of the total mobile source benzene emissions.

Based on the OMS analysis, RVP control, which would be accompanied by a small increase in both benzene content and total aromatic content of gasoline, would have little or no effect on overall fleet emissions or on the number of cancer incidences.[47]

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Several epidemiology studies on workers exposed to benzene have identified benzene as a carcinogen causing leukemia in humans. The upper confidence limit unit risk estimate has been determined from these studies to be 8.0×10^{-6}.[47,48]

4.4 Current and Projected Health Risk

Nationwide exposure levels from both exhaust and evaporative emissions were estimated using the modified NEM exposure model. The speed correction factors at 10 mph and 35 mph required for the NEM inputs were those used for formaldehyde. The nationwide exposure levels predicted by NEM, adjusted upward to account for increased VMT, are 3.09-3.25 ug/m^3 and 1.68-1.77 ug/m^3 for 1985 and 1995, respectively. The range is due to consideration of both a low and high range evaporative emissions estimate for light-duty gasoline-fueled vehicles. Annual cancer incidences from exhaust and evaporative emissions are estimated to be 84-89 in 1985 and 50-52 in 1995. The reason for this marked decrease is the decrease in projected HC in 1995 and, thus, benzene emissions.

Exposure to benzene during refueling includes self-service refueling, occupational exposure (service station attendants) and community exposure in an urban area. Exposure to self-service refueling and occupational exposure was determined by measuring benzene levels in the region of the face of a person refueling a vehicle tank. The exposure in a typical urban area was estimated using a dispersion model.[48] Annual cancer incidences from benzene refueling are estimated to be 8 in 1985 and 7 in 1995.

The total estimated cancer incidences due to mobile source benzene and the contribution of evaporative, exhaust and refueling emissions are given below.

Annual Cancer Incidences due to Mobile Source Benzene

Year	Evaporative*	Exhaust	Refueling	TOTAL
1985	17-22	67	8	92-97
1995	8-10	42	7	57-59

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* Includes low and high range evaporative emissions estimate.

These numbers indicate that, as vehicle hydrocarbon evaporative and exhaust emissions are controlled, the estimated carcinogenic impact due to motor vehicle benzene emissions will decrease. They also indicate that the impact is somewhat less than that predicted for diesel particulates or formaldehyde. However, it should be pointed out that the potency on which these estimates are based is derived from several epidemiology studies and is thus a much firmer potency than that derived for diesel particulates or formaldehyde. The potency for diesel particulates is based on several animal studies rather than human data. For formaldehyde, it is based on a single animal study.

An alternative approach for estimating the exposure and resulting risk of mobile source benzene emissions is to use available ambient monitoring data and assign a mobile source fraction. This approach was used for formaldehyde in order to account for photochemistry.

An urban population weighted average of 10.24 ug/m^3 was calculated based on data from six metropolitan areas. Ambient data for Baltimore, Los Angeles, Northern New Jersey, Philadelphia and Chicago were taken from reference 49. Ambient data for Houston were obtained from reference 46. Their estimated metropolitan area populations in 1983 were taken from reference 15 and used to calculate a population weighted average. Since these cities are in ozone non-attainment areas, the resulting urban concentration may represent more of an upper-bound. A concentration of 7.52 ug/m^3 was selected to represent rural areas.[46]

Since mobile sources appear to be responsible for roughly 85 percent of the total benzene emitted, a mobile source fraction of 0.85 was selected and applied to the estimated urban and rural concentrations above. Resulting urban and rural benzene concentrations due to mobile sources are 8.70 and 6.39 ug/m^3, respectively.

These exposure estimates are then combined with the annual risk estimates to obtain estimates of cancer incidence from current mobile sources. In 1986, the urban and rural risks, using this alternative approach, are estimated to be 179 and 44 cancer incidences, respectively, for a total of 223 cancer incidences. Based on the NEM modeling, emissions of benzene from mobile sources are projected to decrease roughly 40 percent from 1985 to 1995. The mobile source urban, rural and total risk in 1995, accounting for this decrease and the projected population increase, is 116, 29, and 145 cancer incidences, respectively.

These risk estimates are somewhat higher than those calculated with the NEM approach. Both approaches contain uncertainties. For example, temperature effects are only partially accounted for. With the NEM approach, it is assumed that CO and benzene have similar dispersion characteristics. This may not be true. With the ambient apportionment approach, the ambient data may be from fixed site monitors that overrepresent 24-hour exposures of the population. It is also not certain whether the cities chosen are representative of the entire urban population. For the purposes of this report, a range of risk estimates for benzene will be reported, using the results of both approaches. The resulting range of cancer incidences is 92-223 in 1985 and 57-145 in 1995.

4.5 CARB Analysis of Current and Projected Health Risk

In January, 1985, the California Air Resources Board (CARB) identified benzene as a toxic air contaminant. As a result, CARB released a draft report which includes information regarding present (1984) and future (2000) benzene emissions and levels of benzene in California as well as the magnitude of risk posed by benzene emissions from various sources.[50] An addendum to the report was recently prepared.[51] The information in these reports will be briefly summarized in this section. The following section provides a comparison of the EPA and CARB risk estimates. For reference, California contains 11 percent of the U.S. population and 11 percent of the motor vehicles.[19,52]

Total benzene emissions in California were roughly 18,400 tons/year in 1984 and projected to decrease to 13,000 tons/year in the year 2000. The vehicular contribution, which includes both on-road and off-road vehicles as well as other mobile sources, constitute roughly 91 percent of the total emissions in 1984 and 84 percent of the total emissions in 2000. The statewide population in 1984 was 25.8 million and is projected to increase to 31.4 million in 2000.

The population exposure to benzene in California for 1984 and 2000 was estimated using benzene emissions data and population and monitoring data. The estimated statewide annual average exposure to benzene in 1984, 8.4×10^7 ppb-persons, is equal to 25.8 million, the number of people exposed to benzene, times 3.3 ppb, the population weighted annual average benzene concentration. The vehicular contribution (68.1×10^6 ppb-persons) was calculated based on the emissions data. This is similar to the alternative approach described in the previous section. Based on exposure of 25.8 million people, the annual average benzene concentration due to vehicular sources in 1984 is 2.64 ppb (8.5 ug/m^3). This accounts for both urban and rural exposure.

The estimated statewide annual average exposure to benzene in 2000 is 74×10^6 ppb-persons. This was calculated by adjusting the 1984 value to account for the projected reduction in total benzene emissions and the projected increase in population. The vehicular contribution of 63.5×10^6 ppb-persons is again based on the emissions contribution. Based on exposure of 31.4 million people, the annual average benzene concentration due to vehicular sources in 2000 is 2.0 ppb (6.4 ug/m^3). This is a 25 percent reduction relative to 1984.

CARB used a range of risk estimates of 22-170 excess cancers per million people exposed per ppb per 70 years. The range is based on an examination of both mouse and human data. Using this range together with the estimated statewide annual average benzene exposures in 1984 and 2000, the resulting range of risks in 1984 and 2000 are 1,900-14,500 and 1,630-12,600 excess cancer cases in California per 70 years. On an annual basis, this translates to 27-207 cancer cases in 1984 and 23-180 cancer cases in 2000. The vehicular contribution is 21-166 cancer cases in California in 1984 and 20-154 cancer cases in 2000. Expressed as individual risk, the vehicular contribution is 8.11 - 64.3 x 10^{-7} in 1984 and 6.4 - 49.0 x 10^{-7} in 2000.

4.6 Comparison of EPA and CARB Health Risks

The range of cancer incidences given in this report is 92-223 in 1986 and 57-145 in 1995. In order to compare these risk estimates to those of CARB, they are expressed as individual risks. The resulting individual risk estimates are 3.8 - 9.3 x 10^{-7} in 1986 and 2.2 - 5.6 x 10^{-7} in 1995. The upper end of these risk estimates are roughly equivalent to the lower end of the CARB estimates.

CARB used a range of risk estimates of 22-170 excess cancers per million people exposed per ppb per 70 years. In terms of unit risks, this translates to 6.81 - 52.6 x 10^{-6}. The upper confidence limit unit risk used in this study is 8 x 10^{-6}; this is near the low end of the range of CARB unit risks. CARB's high end risk estimate is based on animal studies, whereas EPA's risk estimate is based on human studies.

The CARB estimate of the benzene concentration due to vehicular sources in 1984 is 8.5 ug/m^3. In comparison, the range of estimated exposures in this study for 1985 is 3.2 - 8.1 ug/m^3 (accounting for both urban and rural exposure, as CARB has done). Clearly, the CARB estimate exceeds the exposure estimates used in this study. This is not surprising since monitoring data indicate that benzene concentrations in urban areas of California are higher than most other areas of the country. (The high benzene level in Los Angeles was factored into the national estimate used in this study.)

The CARB projections do not show benzene emissions dropping as rapidly as predicted in this report. CARB predicts that vehicular benzene emissions will decrease 35 percent from 1984 to 2000. This can be compared to a 40 percent decrease from 1985 to 1995 given in this report. Also, when the CARB risk is expressed in terms of total cancer cases, the number of cancer cases only decreases by 13 percent from 1984 to 2000.

This is because the decrease in emissions is slightly offset by the projected increase in population. The population in California is projected to increase 22 percent from 1984 to 2000. In comparison, the U.S. population is projected to increase 8 percent from 1986 to 1995.

When reviewing the CARB risk estimates, it should also be remembered that the vehicular contribution includes on-road vehicles, off-road vehicles, trains, ships, aircraft, mobile equipment and utility equipment. The EPA estimates only include emissions from on-road vehicles. Most of the difference seen in the two estimates, however, is attributed to the wide range of unit risks used by CARB.

4.7 Current Activities

The California Air Resources Board (CARB) is considering implementing regulations requiring control of motor vehicle benzene emissions. This may include a limit on the benzene content of gasoline and/or stricter light-duty exhaust HC standards. The first measures being considered are a more stringent exhaust HC standard by the end of 1987, and possible changes in the evaporative test procedure, e.g., multiple diurnal tests, longer soak times, and higher soak temperatures.

The EPA Office of Air Quality Planning and Standards (OAQPS) has designated benzene as a hazardous air pollutant under Section 112 of the Clean Air Act and is implementing necessary controls for stationary sources. The EPA Office of Mobile Sources (OMS) is determining if any motor vehicle controls specific to benzene are needed. Also, OMS is determining how other regulations that may be proposed for additional hydrocarbon control would affect benzene.

5.0 GASOLINE VAPORS

Several years ago, the American Petroleum Institute released the results of a lengthy animal inhalation study showing that totally vaporized gasoline resulted in an increase in kidney tumors in male rats and liver tumors in female mice.[53] EPA evaluated these data in depth to determine the potential carcinogenic impact.[48,54] From these studies, EPA calculated the following plausible upper limit carcinogenic potencies for exposure to 1 ppm for a year. The maximum likelihood potency is lower by almost a factor of two for both data sets.

Gasoline Carcinogenic Potencies Based on API Study

Rat data	4.9×10^{-5}/ppm
Mice data	2.9×10^{-5}/ppm

Exposure to gasoline vapors and benzene from gasoline during refueling was estimated based on an American Petroleum Institute study that involved measuring gasoline and vapor levels in the region of the face of a person refueling a vehicle tank. The exposure in a typical urban area for these refueling emissions was also estimated by using the Industrial Source Complex (ISC) dispersion model to calculate annual concentrations.[48]

The exposure numbers determined were combined with the rat potency data to calculate the potential carcinogenic impact due to refueling emissions. These numbers for both benzene and gasoline vapors are as below.

Potential Carcinogenic Impact of Refueling Loss Emissions

Exposure	Annual Incidences Benzene	Gasoline Vapors
Self-service refueling	5	35
Occupational exposure (service station attendants)	2	17
Community exposure in an urban area	0.5	13
TOTAL EXPOSURE	7.5	65

There are several limitations to the use of the API bioassay, however. First, totally vaporized gasoline was used which includes higher molecular weight components as well as the lighter components. Some short term animal tests done by API indicate that the branched chain paraffins (e.g., C-6, C-7, C-8) are the ones that would cause kidney damage. It is not certain what relation the kidney damage has to the kidney tumors in the rats. However, it is known that most (i.e., 80-90%) of the gasoline vapors in a realistic refueling episode are below C-6 while only about 23% of the vaporized gasoline is below C-6.[55,56]

Another limitation to the API work is the apparent involvement of a low molecular weight protein (alpha 2-globulin) that is present only in rats. It is thought that this protein, which is synthesized in the liver of mature, male rats, accumulates in the kidney following hydrocarbon exposure. The effect results in kidney damage (nephropathy, a chronic inflammation and vascular thickening of portions of the kidney) similar to that known as "old rat nephropathy". This condition frequently occurs spontaneously in older rats; the presence of hydrocarbons accelerates this change. There is some concern that this kidney condition is related to the small cancerous growths found. Alpha 2-globulin is not present in humans which are not affected by this kidney condition. Female mice have a large and variable rate of spontaneous liver tumors. Like the case of the rats, some questions have been raised about extrapolating the results with the mice to humans. These issues have been discussed in a report by the Health Effects Institute (HEI) which concluded the following.

> ...the usefulness of available animal and human data in helping to determine health risks is quite limited. Unburnt gasoline vapors may, upon further investigation, prove to present significant carcinogenic risks for humans. The evidence is not available to make that statement today. Significant additional research would have to be undertaken to understand important mechanisms of action, physiological differences between test animals and people and the extent and nature of exposures.[57]

It should be noted that HEI has since decided to perform no research in the gasoline vapor area. Also, aside from some limited work being done by API, EPA is aware of no additional health research being done on gasoline vapors.

In an internal EPA memo, EPA has concluded the following about the potential carcinogenicity of gasoline vapors and the EPA risk assessment discussed above.

> All issues raised by HEI were known to EPA and thoroughly assessed in EPA's analyses of the same issues. EPA's quantitative risk assessment for carcinogenicity of unleaded gasoline vapors was prepared according to EPA Guidelines and expresses its uncertainties, which are not greater than EPA routinely addresses. Further research would not change the underlying evidence of the carcinogenicity of gasoline vapors but may shed light on which components are responsible for the carcinogenic response and this could alter the risk calculation.[58]

On an overall basis then, it appears that gasoline vapors must be regarded as a potential human carcinogen. Also, gasoline vapors represent a hydrocarbon source which may need to be controlled to help attain the ozone National Ambient Air Quality Standard. EPA has not yet made a decision to propose controls for gasoline vapors or a decision on the form of the control.

6.0 GAS PHASE ORGANICS

6.1 Formation and Control Technology

Gas phase organics, or volatile organic compounds (VOC), are present in both exhaust and evaporative emissions. Over 300 gas phase organics have been identified.[59] The majority of VOC are formed from incomplete combustion of the fuel while others are simply evaporated fuel components. The emphasis of this section will be on exhaust compounds; evaporative compounds are covered in Section 5 discussing gasoline vapors.

The total mass of VOC has decreased since 1975 due to introduction of the oxidation catalyst (caused by more stringent HC standards). More advanced control technology (i.e., 3-way and 3-way plus oxidation catalysts) introduced in the early 1980's to allow further simultaneous control of NOx and HC has reduced the total mass even further.

6.2 Composition

The majority of gas phase organics consist of unsaturated and saturated hydrocarbons along with benzene, alkyl benzenes, aliphatic aldehydes and a variety of polycyclic aromatic hydrocarbons (PAH) including nitro-PAH. A list of the VOC measured in ECTD contractor studies (that include unregulated emissions characterization) is given in Table 6-1. It should be noted that this is not meant to be a complete list; instead, it represents those compounds for which the most emissions data are available.

Studies have been conducted to determine the mass and the detailed hydrocarbon composition of vehicle exhaust.[30,60] Gas chromatographic (GC) analysis was used in these studies to identify individual hydrocarbons of carbon numbers 1 through 10 (C_1 - C_{10}). In a recent study, 82 individual hydrocarbons and 10 aldehydes were measured in the exhaust of 46 in-use 1975-1982 gasoline-fueled vehicles.[30] The gas chromatographic conditions employed did not permit identification of each individual compound above C_{10}. Fortunately, the hydrocarbon composition of gasoline engine exhaust consists primarily of components with carbon numbers 1 through 10 so a fairly complete description of VOC emissions from gasoline-fueled vehicles is possible with GC analysis. The exception is the small quantity of PAH which is also present in gasoline exhaust. A more detailed discussion of gas phase PAH will be presented later in this section.

Unlike gasoline-fueled vehicles, the VOC emitted from diesel-fueled vehicles range from C_1 to about C_{40}, with the majority being below C_{25}. The C_1-C_{10} hydrocarbons result almost entirely from the combustion process, which involves

Table 6-1

VOC Unregulated Emissions Most Commonly Characterized

Total Hydrocarbons

Individual Hydrocarbons

Methane
Ethylene
Ethane
Acetylene
Propane
Propylene
Benzene
Toluene

Aldehydes and Ketones

Formaldehyde
Acetaldehyde
Acetone
Isobutyraldehyde
Methyl Ethyl Ketone
Crotonaldehyde
Hexanaldehyde
Benzaldehyde

cracking, and possibly subsequent polymerization, of higher molecular weight materials. It is postulated that the C_{10}-C_{25} hydrocarbons result, to a large extent, from uncombusted fuel, and the C_{15}-C_{40} hydrocarbons from lubricants.[4]

Gas phase PAH have been characterized in the exhaust of both gasoline- and diesel-fueled vehicles.[61,6] Some of the PAH identified included anthracene and phenanthrene, fluoranthene, pyrene, benzo(a)anthracene and isomers, chrysene, indenopyrene, benzo(ghi)perylene, benzo(e)pyrene, benzo(a)pyrene and coronene. These compounds, which contain two to six benzene rings, have also been identified in diesel particulate extracts. With the exception of anthracene, phenanthrene and possibly pyrene (the data for pyrene appear conflicting), the majority of these PAH are emitted in the particulate phase. This is particularly true for benzo(a)pyrene. Particulate/gas phase mass ratios for benzo(a)pyrene range from roughly 4:1 [61] to 15-27:1.[6]

6.3 Mutagenicity of VOC

Two studies were conducted to determine the relative mutagenicity of the gas and particulate phases in the exhaust of gasoline and diesel-fueled vehicles. A XAD-2 trapping system was used in both studies to collect the gas phase compounds (greater than C_7). The highly volatile compounds, i.e., less than C_7, can not be collected on a XAD-2 trap. Mutagenicity of the trapped compounds was determined with the Ames test (Salmonella typhimurium).

In the first study, conducted with gasoline-fueled stratified charge engine, only 3 percent of the total direct-acting (i.e., mutagenicity decreases with the addition of S9 activation) and 5 percent of the total indirect-acting mutagenicity was found in the gas phase sample.[6] Most of the gas phase indirect-acting mutagenicity was found in the non-polar fraction which may be due to the presence of PAH.[6]

In the second study, similar results were found.[62] The mutagenic activity of the gas phase emissions from the three gasoline-fueled vehicles tested were at or near background levels. For the single diesel-fueled vehicle tested, the mutagenic activity of the gas phase emissions, expressed as revertants per mile, was less than 11 percent of the mutagenicity of the particle-bound organics.

6.4 Risk Associated with Individual VOC

For the purpose of this report, available data on detailed hydrocarbon emissions were reviewed, and individual compounds were selected which have unit risks associated with them. Of the gas phase organics emitted in motor vehicle exhaust, benzene, formaldehyde, 1,3-butadiene, ethylene and benzo(a)pyrene have unit risks. Benzene and formaldehyde have been discussed in previous sections.

1,3-Butadiene

It is difficult due to the limited data available to develop accurate NEM inputs for 1,3-butadiene. Only data for light-duty gasoline-fueled vehicles exist. 1,3-Butadiene poses an additional problem because 1,3-butadiene and n-butane coelute and thus have the same retention point on the gas chromatograph. Emission characterization studies to date have not attempted to determine the percentage of the peak due to 1,3-butadiene. Therefore, assumptions must be made about the percentage each compound contributes to this peak. In a previous study, it was assumed that 20 percent of the peak was due to 1,3-butadiene, for the purpose of grouping the detailed HC data into various compound classes.[63] Based on data collected in the Lincoln Tunnel, 1,3-butadiene constitutes 13.9 percent of the total peak, although unknown dilution with air containing n-butane and 1,3-butadiene complicates the analysis.[64] Morning ambient samples, probably daily maximums, collected in 1984 and 1985 show much lower percentages, with overall averages of 5.08 and 4.21 percent, respectively.[64] For this report, fifteen percent was chosen as an upper limit.

1,3-Butadiene emissions were expressed as a percentage of the total exhaust HC predicted by MOBILE3. Based on the data from 46 in-use gasoline-fueled vehicles provided in reference 30 (and the fifteen percent assumption), 1,3-butadiene is roughly 0.94 percent of the total FID exhaust HC. Due to the lack of data for the other vehicle classes, this percentage was simply applied to the MOBILE3 composite exhaust HC emission factor. It was further assumed that the percentage would remain the same from 1986 to 1995. Composite 1,3-butadiene emission factors for 1986 and 1995 are 0.0238-0.0263 g/mile and 0.0121-0.0143 g/mile, respectively. The range accounts for both the presence and absence of an Inspection/Maintenance program. The speed correction factors were those used for formaldehyde.

The modified NEM model was used to estimate exposures. Nationwide urban and rural exposure in 1986 is estimated to be 0.69-0.76 ug/m^3 and 0.32-0.35 ug/m^3, respectively. In 1995, nationwide urban and rural exposure is estimated to be 0.42-0.49 ug/m^3 and 0.19-0.23 ug/m^3, respectively.

These exposure estimates are for direct emissions of 1,3-butadiene and do not account for reactions of 1,3-butadiene in the atmosphere. Available ambient monitoring data were reviewed and compared to the exposure estimates. A condensed summary of 1,3-butadiene monitoring data is given in Table 6-2.[64] As seen in the table, average mean values in urban settings range from 0.77-24.23 ug/m³. The 1986 NEM estimate of urban exposure from motor vehicles (0.69-0.76 ug/m³) lies near the low end of the range. The monitoring data indicate that 1,3-butadiene is, in fact, emitted from motor vehicles as evidenced by the elevated levels in tunnels. The average mean value in a rural setting (0.67 ug/m³) is also within the same order of magnitude as the NEM rural estimate (0.32-0.35 ug/m³).

More recent 1986 1,3-butadiene monitoring data are now available for 18 cities.[65] These data represent 6-9 a.m. averages of about 10-15 samples for each city, measured during the summer months. The average concentrations range from 0.24-1.98 ug/m³ (Data for Los Angeles were not obtained). Again, the 1986 NEM estimate of urban exposure from motor vehicles lies within this range.

The EPA estimate for the upper confidence limit unit risk for 1,3-butadiene has changed substantially. Based on inhalation studies of 1,3-butadiene in mice, a 95 percent upper confidence limit unit risk for 1,3-butadiene is estimated to be 2.8×10^{-4}.[66] This is much greater than the previous estimated unit risk of 4.6×10^{-7}.

Estimates of cancer incidence for 1986 and 1995 are given in Table 6-3. The total risk in 1986 (with the fifteen percent assumption) ranges from 593-656 cancer incidences and drops to 391-460 cancer incidences in 1995. Preliminary emission characterization results indicate the presence of 1,3-butadiene, but the amount has not yet been quantified. Therefore, a lower risk estimate of zero will also be used. The resulting ranges of cancer incidences for 1986 and 1995 given in Table 6-3 are 0-656 and 0-460, respectively.

Ethylene

The upper confidence limit unit risk of 2.7×10^{-6} for ethylene was provided in the Six Month Study although it was not developed by EPA. Unlike 1,3-butadiene, more extensive and reliable emissions data exist to construct NEM inputs and predict resulting exposure and risk from direct emissions of ethylene. Ethylene was handled in much the same way as formaldehyde. Emissions were expressed as a percentage of the total exhaust HC predicted by MOBILE3. Ethylene in evaporative emissions is negligible and was not considered.

Ethylene emissions, expressed as a percentage of total hydrocarbons, for the vehicle classes in 1986 and 1995 are given below:

Table 6-2

1,3-Butadiene Monitoring Data: Condensed Summary

Location	ug/m^3 Average Mean Value	Maximum Value	Comments
Houston, TX area*	5.22 2.81	88.61 33.76	incl. tunnels excl. tunnels
Los Angeles, CA area*	24.23	88.39	
Riverside, CA	3.98	6.19	
Atlanta, GA	5.57		
Lincoln Tunnel, NY	20.11 1.64	24.09 2.71	in tunnel outside air
Columbus, OH	0.77	5.47	
Denver, CO	1.7	7.6	only one sample set
Jones State Forest	0.67	2.41	non-urban setting

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* All locations within approximately 15 miles of the metropolitan area.

Table 6-3

Annual Cancer Risk from Direct Emissions of 1,3-Butadiene from Mobile Sources

	Urban	Rural	Total
1986			
With I/M	0-514	0-79	0-593
Without I/M	0-568	0-88	0-656
1995			
With I/M	0-339	0-52	0-391
Without I/M	0-399	0-61	0-460

Vehicle Class	% of Total Exhaust HC 1986	 1995
LDGV	9.0	6.4
LDGT 1,2	9.0	6.4
LDDV	11.0	11.0
LDDT	11.0	11.0
HDGV	13.0	13.0
HDDV	9.0	9.0

Data were taken from references 30, 32-34, 36, 38, and 40. The percentages of exhaust HC for the light-duty gasoline classes were projected to decrease from 1986 to 1995 due to the increasing penetration of 3-way and 3-way plus oxidation catalyst-equipped vehicles and trucks. The speed correction factors used to compute 10 mph and 35 mph emission factors were those used for formaldehyde. Composite FTP emission factors for 1986 and 1995, with and without I/M, are 0.2367-0.2607 g/mile and 0.0946-0.1092 g/mile, respectively.

Estimates of cancer incidence for 1986 and 1995 are given in Table 6-4. As seen in this table, the total risk in 1986 ranges from 55-60 cancer incidences and drops to 29-31 cancer incidences in 1995.

Two important limitations need to be mentioned when discussing the risk estimates for ethylene. The first is the fact that ethylene is photochemically reactive. The risk estimates are for direct emissions of ethylene and do not account for reactions of ethylene in the atmosphere.

The second, and most important, limitation is the unit risk estimate. The unit risk quoted in the Six Month Study was estimated by Clement Associates, Inc. for EPA but has not been endorsed by EPA.[67] There is no available evidence that ethylene is carcinogenic although ethylene oxide, a metabolite of ethylene, has been shown to be an animal carcinogen. EPA has calculated a unit risk for ethylene oxide. Clement assumed a downward difference in potency of 100 to calculate a unit risk for ethylene. The basis for this assumed difference in potency was not given. Since there is no available evidence that ethylene is carcinogenic, the risk estimates must be regarded as extremely tentative. For this reason, a lower risk estimate of zero will also be used.

Benzo (a) Pyrene (B(a)P)

Gas phase B(a)P is emitted in small quantities. The risk posed by gas phase B(a)P will be considered together with particle-associated B(a)P. It will be assumed that the particle-associated B(a)P emission factor used will adequately represent what little gas phase B(a)P may also be present. The risk from particle-associated B(a)P is discussed in Section 7.

Table 6-4

Annual Cancer Risk from Direct Emissions Of Ethylene from Mobile Sources*

	Urban	Rural	Total
1986			
With I/M	0-47	0-8	0-55
Without I/M	0-52	0-8	0-60
1985			
With I/M	0-25	0-4	0-29
Without I/M	0-27	0-4	0-31

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* Lower limit of zero used due to the uncertainty of the unit risk estimate.

6.5 Reactivity of VOC

The atmospheric photochemical reaction products of mobile source volatile organic compounds (VOC) are largely unknown. It is likely that carcinogens may be formed after emissions leave the vehicle as well as some carcinogens degrading to non-carcinogenic compounds. A study was conducted by Calspan Corporation for the Coordinating Research Council, the objective being to investigate the fate of diesel exhaust in the atmosphere.[68] Smog chamber experiments with diluted diesel exhaust were conducted to investigate parameters such as the presence or absence of UV irradiation, dilution ratio, aging and exposure to ozone and NO_2. The presence of ozone increased the mutagenic response of the collected particle-bound organics, in some cases by as much as an order of magnitude. Similar results were obtained when NO_2 and/or irradiation is present in the chamber. Thus, it appears that the mutagenicity of diesel particle-bound organics is affected by ambient conditions.

It also appears likely that irradiation of even innocuous VOC compounds may lead to the formation of mutagens. In one study, irradiation of propylene, SO_2 and NO_x did demonstrate a mutagenic response. [69]

Mobile source VOC is known to contain photochemically reactive compounds. The percent composition of exhaust hydrocarbons for pre-1975, 1975-1980 and 1981-1982 model year gasoline-fueled vehicles is given in Table 6-5.[30,63] The table provides a general indication of changes in reactivity with advancing control technology. Pre-1975 vehicles are not catalyst-equipped. Model year 1975-1980 vehicles are primarily equipped with oxidation catalysts whereas 1981-1982 vehicles are primarily equipped with 3-way and 3-way plus oxidation catalysts.

Olefins and aromatics are all fairly reactive; paraffins and acetylene are less reactive. In particular, methane is essentially non-reactive. From Table 6-5, it can be seen that vehicles equipped with catalysts, particularly 3-way and 3-way plus oxidation catalysts, emit a higher percentage of methane in their exhaust than do non-catalyst-equipped vehicles. Most catalytic converter systems preferentially oxidize non-methane hydrocarbons because methane is harder for the catalytic converter to oxidize. Since methane is essentially non-reactive, the total photochemical reactivity of the HC mixture tends to be reduced by the catalyst. In addition, the catalyst reduces the total hydrocarbon mass and generally oxidizes the unsaturated HC compounds to a greater extent than the saturated compounds. This is evidenced by the decreasing percentages of olefins and aromatics coupled with the increasing percentage of paraffins in the exhaust.

Table 6-5

Percent Composition of Exhaust Hydrocarbons

	% of Total Exhaust HC by Model Year		
	Pre-1975*	1975-1980**	1981-1982**
Paraffins (Methane)	41.0 (9.2)	49.2 (9.3)	66.5 (21.8)
Olefins	30.0	23.2	15.2
Acetylene	8.8	3.4	0.9
Aromatics	20.4	25.1	17.4

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* Reference 63
** Reference 30

Aldehydes were not included in Table 6-5. Aldehydes, particularly formaldehyde, are photochemically reactive. The catalyst has also been found to reduce formaldehyde and total aldehyde emissions. Thus, it appears that, as non-catalyst-equipped vehicles are phased out of the fleet, the reactivity of vehicle exhaust will decrease. The reaction products of mobile source VOC to which people are exposed remains to be determined.

6.6 Current Activities

EPA-ORD is conducting a long-term research project referred to as the Integrated Air Cancer Project (IACP). The goal of the IACP is to identify the principal airborne carcinogens and their sources. Initially, airsheds with one or two emission sources will be examined, followed by areas of increasing complexity.

In Phase I, initial field measurements were conducted in Albuquerque, New Mexico and Raleigh, North Carolina. The purpose of Phase I was to evaluate and select the sampling, analytical and bioassay methodologies, as well as other approaches for an integrated field study.

In Phase II, which will be conducted in FY86 and FY87, an integrated field study will be conducted in Boise, Idaho. The two major emission sources will be mobile sources and emissions from wood combustion for residential heating. Some of the objectives are to: 1) identify and quantify classes of compounds in the ambient air resulting from residential wood combustion and motor vehicles, 2) quantify the relative contributions of these sources to the mutagenic activity, organic and fine particulate mass of ambient airborne pollutants, and 3) characterize the chemical changes which may occur to the source emissions in the atmosphere and assess the resultant changes in mutagenic response.

In conjunction with the integrated field study in Boise, separate smog chamber studies will be conducted under IACP to assess the mutagenicity of vehicle exhaust before and after irradiation. These results could provide at least an initial indication of the photochemical transformation of mobile source emissions, both gas phase and particle-associated, and the resulting effect on mutagenicity.

7.0 ORGANICS ASSOCIATED WITH NON-DIESEL PARTICULATE

7.1 Emission Rates and Composition

This section will deal with the organic compounds associated with gasoline particulate emissions.

Table 7-1 presents total particulate and associated soluble organic fraction (SOF) emissions for the different vehicle classes.[2,70] Gasoline-fueled vehicles emit far less particulate than their diesel counterparts. As discussed in Section 2.1, it is thought that nitro-polycyclic aromatic hydrocarbons (PAH) such as nitropyrenes, dinitropyrenes and nitrohydroxypyrenes together account for much of the mutagenicity of diesel particulate emissions. Particulate emissions from gasoline-fueled vehicles contain significantly less of these nitro-PAH's; however, as seen in Table 7-1, the mutagenicity of gasoline SOF, expressed as reverents/ug SOF, is greater than diesel SOF. Also, unlike diesel SOF, the mutagenic activity of gasoline SOF increases with the addition of S9 activation, indicating indirect-acting activity. This suggests that the classical PAH's may be responsible for the mutagenicity of gasoline SOF, rather than the nitro-PAH's.

7.2 Risk from Non-Diesel Particle-Associated Organics

For gasoline-fueled vehicles, three different approaches were taken to estimate the risk from non-diesel particle-associated organics, referred to here as gasoline particle-bound PIC (products of incomplete combustion). The first approach estimates the risk of B(a)P emissions from gasoline-fueled vehicles and assumes no risk from the remaining gasoline particle-bound PIC emissions. The second approach uses B(a)P emissions as a surrograte for gasoline particle-bound PIC emissions. Unlike the first approach, which uses the unit risk for B(a)P, the second approach uses the PIC unit risk presented in the Six Month Study. The third approach uses estimated gasoline particle-associated organic emission rates together with a unit risk for gasoline particle-associated organics. All three approaches and resulting risk estimates will be described in this section.

The first approach uses B(a)P emission factors from gasoline-fueled vehicles together with the B(a)P unit risk. The annual cancer risk of B(a)P from gasoline-fueled vehicles was determined by multiplying the B(a)P risk obtained in the Six Month Study by the ratio of the emission factors (this

Table 7-1

Particulate Emissions and Mutagenicity*

	Leaded LDGV	Unleaded LDGV	Leaded HDG	LDD	HDD
Total particulate, mg/mile	102.5	31.7	735	606.8	1948
SOF, mg/mile	21.1	14.4	27.6	124.1	385.2
SOF, as % of total particulate	20.6	45.4	3.8	20.5	19.8
Benzo(a)pyrene, ug/mile	14.6	3.2	39.5	4.5	2.3
Nitropyrene, ug/mi	0.20	0.24		7.4	
TA98, -S9, rev/ug SOF**	7.3	7.6	5.3	4.1	0.88
TA98, +S9, rev/ug SOF	12.5	13.4	16.1		0.84
TA98, -S9, rev/mi ($x10^3$)	152	42.1	110.5	509	287.4
TA98, +S9, rev/mi ($x10^3$)	258	79.3	428.3		279.4

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* For the light-duty vehicles, emissions were collected during the FTP; for heavy-duty vehicles, the transient test procedure was used.

** Denotes revertants per microgram of the soluble organic fraction (SOF), using Salmonella typhimurium strain TA98 with or without metabolic activation (S9).

study/Six Month Study).* This approach assumes that there is no risk from the remaining gasoline particle-bound PIC.

The B(a)P emission factors were taken from references 2 and 70 and summarized in Table 7-1. B(a)P emission factors can vary substantially from one reference to another. The emission factors used represent the higher end of the range. Since gas phase B(a)P is emitted in such small quantities, it is assumed that the emission factors used adequately represent what little gas phase B(a)P may also be present. The unleaded and leaded LDGV weightings were 0.88 and 0.12, respectively to compute a composite LDGV emission factor. These weightings are based on MOBILE3 estimates for 1986. The resulting overall composite emission factor is 5.56×10^{-6} g/mile. The exclusion of diesel-fueled vehicles does not have much impact on the composite emission factor since most of the B(a)P is emitted from gasoline-fueled vehicles.

The composite emission factor of 5.56×10^{-6} g/mile is roughly one-third of the emission factor used in the Six Month Study (1.66×10^{-5}g/mile). The primary reason for this difference is that the Six Month Study assumed a 50/50 split for leaded and unleaded gasoline-fueled vehicles. A B(a)P unit risk of 3.3×10^{-3} was used for both the Six Month Study and this approach.

The B(a)P risk from motor vehicles estimated in the Six Month Study is 0.02 cancer incidences per urban million. The current (1986) cancer risk of B(a)P from gasoline-fueled vehicles using the first approach was determined by simply multiplying the risk obtained in the Six Month Study by the ratio of the composite emission factors ($5.56 \times 10^{-6}/1.66 \times 10^{-5} = 0.33$). The resulting annual cancer risk is 0.007 per urban million, or 1.3 cancer incidences, assuming an urban population of 180 million in 1986.

In 1995, virtually all LDGV and HDGV will be using unleaded fuel. The composite B(a)P emission factor will decrease as a result to roughly 2.95×10^{-6} g/mile. When this emission factor is used, the resulting cancer risk is 0.004 per urban million, or 0.78 cancer incidences, assuming an urban population of 195 million in 1995.

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* This is the first place in the report where the Six Month Study exposures are used in place of NEM-predicted exposures. The Six Month Study used the GAMS dispersion model developed by EPA's Office of Toxic Substances to calculate exposures. Area sources were generally assumed to be distributed equally throughout each county. As a very crude comparison, assuming the same FTP emission factor, the NEM-predicted exposure is roughly 1.6 times as high as that predicted by GAMS.

The second approach uses B(a)P emission factors from gasoline-fueled vehicles together with the PIC unit risk (which is expressed per unit of exposure of B(a)P) to estimate the annual cancer risk of PIC from gasoline-fueled vehicles. The PIC unit risk is 4.2×10^{-1}. The Six Month Study also used B(a)P emission factors together with the PIC unit risk to estimate the PIC risk from motor vehicles. The annual cancer risk of PIC from gasoline-fueled vehicles was determined by multiplying the PIC risk obtained in the Six Month Study by the ratio of the B(a)P emission factors.

The PIC risk from motor vehicles estimated in the Six Month Study is 2.07 per urban million. When this is multiplied by the ratio of the B(a)P emission factors ($5.56 \times 10^{-5} / 1.66 \times 10^{-5} = 0.33$), the resulting annual cancer risk is 0.68 per urban million, or 122 cancer incidences in 1986. When the estimated 1995 B(a)P emission factor is used (2.95×10^{-6}), the resulting cancer risk is 0.37 per urban million, or 72 cancer incidences in 1995.

The third approach uses estimated emission rates of gasoline particle-associated organics as an unspeciated mixture together with a unit risk for these organics. Exposures were estimated using the modified NEM model.

Emission rates of gasoline particle-associated organics in 1986 and 1995 were estimated using the methodology in reference 71. Unfortunately, the organic emission factors contained in reference 71 include both soluble organics and elemental carbon. The organic emission factors in reference 71 were revised to include only the soluble organic fraction (SOF). The SOF emission factors used for the various vehicle classes are given in Table 7-2. SOF emission factors for light-duty gasoline-fueled vehicles and trucks were taken from references 70 and 72. SOF emission factors for the heavy-duty gasoline-fueled trucks were taken from references 73, 74 and 75. The pre-1987 heavy-duty data were obtained using EPA's transient chassis test procedure. It is assumed that the more stringent emission standards for 1987 and later heavy-duty gasoline vehicles in the 8,501-14,000 lbs range (classes 2B and 3) will require the use of catalysts. The SOF emission factors in Table 7-2 differ somewhat from the emission factors in Table 7-1. The reasons for this difference are: 1) the emission factors in Table 7-2 are specific by model year unlike those in Table 7-1, and 2) more references were used in compiling Table 7-2.

MOBILE3 VMT fractions for 1986 and 1995 for the various vehicle classes were used to calculate composite emission factors. Composite emission factors were estimated assuming both the presence and absence of an Inspection/Maintenance (I/M) program. (The effect of the I/M program is to reduce the rates of misfueling and catalyst removal, which affect particle-bound organic emission rates.) An FTP average speed

Table 7-2

Gasoline Particle-Associated Organic Emission Factors

Light-Duty Gasoline Vehicles and Trucks

Model Year	Control System	Fuel Used	Emission Factor grams per mile
Pre-1970	NOCAT	Leaded	0.087[a]
1970-1974	NOCAT	Leaded	0.023[b]
1975+	NOCAT	Leaded	0.019[b]
1975+	CAT	Unleaded	0.005[b]
1975+	CAT	Leaded	0.023[c]
1975+	NOCAT	Unleaded	0.019[d]

Heavy-Duty Gasoline Trucks

Model Year	Control System	Fuel Used	Emission Factor grams per mile
Pre-1987	NOCAT	Leaded	0.072[e]
1987+	CAT	Unleaded	0.013[f]
1987+	CAT	Leaded	0.050[g]
1987+	NOCAT	Leaded	0.105[h]

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[a] From reference 72

[b] From reference 70

[c] Same as 1970-1974 no catalyst leaded value.

[d] Same as 1975+ no catalyst leaded value.

[e] From references 73, 74 and 75. The average class 2B truck value was assigned a weighting of 60% based on VMT.

[f] Average class 2B no catalyst leaded truck value multiplied by ratio of light-duty 1975+ catalyst unleaded and 1975+ no catalyst leaded values (0.050 x 0.005/0.019).

[g] Average class 2B no catalyst leaded truck value.

[h] Average no catalyst leaded truck value for classes 5 and 6.

of 19.6 mph was assumed. The estimated fleet composite emission factors are given below.

Gasoline Particle-Associated Organic Emissions (g/mile)

	With I/M	Without I/M
1986	0.0075	0.0082
1995	0.0048	0.0058

Based on data obtained over a number of different driving cycles, no trends are apparent with regard to speed.[70] Therefore, the g/mile emission factors above were used to calculate g/min emission factors at 10 mph, 19.6 mph and 35 mph, as required by the model. Idle emission factors were also determined based on data for non-catalyst-equipped and oxidation catalyst-equipped vehicles.[40] Idle emission factors for 1986 and 1995 are 0.0196 g/min and 0.0138 g/min, respectively.

A unit risk estimate for gasoline particle-associated organics was estimated by EPA scientists in 1983.[7] Like diesel particulate, the unit risk estimate has not been reviewed by the Science Advisory Board (SAB) and an official EPA risk assessment has not been done. It is based on data for only one catalyst-equipped vehicle. Furthermore, the vehicle had exceptionally high exhaust emissions, comparable to those from a non-catalyst-equipped vehicle. It was originally chosen on this basis since it was easier to collect enough extractable organics for analysis. The mutagenic activity of the particle-associated organics from this vehicle, as indicated by the Ames Salmonella strain TA-98 bioassay, is on the low end of the range, when compared with other catalyst-equipped vehicles. As a result, the vehicle should be considered to be of uncertain representativeness. An upper confidence limit unit risk estimate based on this vehicle is 2.5×10^{-4}. The bioassays used to estimate the unit risk were the same as those used to estimate the unit risk for diesel particulate, and the same approach was used.

Nationwide urban and rural exposure in 1986, using the modified NEM, is estimated to be 0.20-0.21 ug/m^3 and 0.17-0.19 ug/m^3, respectively. The range accounts for both the presence and absence of an Inspection/Maintenance program. In 1995, nationwide urban and rural exposure is estimated to be 0.13-0.15 ug/m^3 and 0.11-0.13 ug/m^3, respectively.

Estimates of lung cancer incidence for 1986 and 1995 are given in Table 7-3. The total risk in 1986 ranges from 163-176 cancer incidences and drops to 115-136 cancer incidences in 1995.

For this report, a range of risk estimates for gasoline PIC will be reported, which encompasses the results of all three approaches. The resulting range of cancer incidences is 1.3-176 in 1986 and 0.78-136 in 1995.

Table 7-3

Annual Lung Cancer Risk from Gasoline Particle-Associated Organics

	Urban	Rural	Total
1986			
With I/M	1.3-127	0-36	1.3-163
Without I/M	1.3-136	0-40	1.3-176
1995			
With I/M	0.78-90	0-25	0.78-115
Without I/M	0.78-106	0-30	0.78-136

8.0 DIOXINS

8.1 Composition

Over 75 different chlorinated dioxin isomers have been identified. One of the 22 isomers with four chlorine atoms is 2,3,7,8-tetrachlorodibenzo-p-dioxin (2378-TCDD). This dioxin compound has a high molecular weight and exists in the particulate state or is adsorbed onto particulates. It is the dioxin compound of most interest since it is thought to be the most toxic of the chlorinated dioxins and is most often associated with exposure and potential health risks to humans based on available data.

8.2 Emissions

Some qualitative analytical measurements have found dioxin to be present in the muffler scrapings of vehicles using either leaded or unleaded gasoline. Also, some measurements have shown this compound to be present in diesel particulate. Some work has been done attempting to measure this compound quantitatively in vehicle exhaust. Since it is apparently emitted in only trace quantities (e.g., 10^{-9} g/mile), it is very difficult to collect enough particulate sample for analysis.[76] Also, a radioactive tracer compound must be used to correct for losses in sample work-up.

8.3 Concentrations of Dioxins

Most reports of environmental contamination from dioxins have concerned non-air releases of dioxin from industrial or chemical processes (e.g., pesticide production) or measurement of dioxin levels in fish in waters that could have been contaminated by non-air routes. At this point, there are no accurate measurements of ambient levels of dioxin since it would be present in such low levels in the ambient air. Thus, it is not possible to say for certain whether the known sources of dioxin such as municipal and industrial incinerators account for most of the dioxin found in the ambient air although it is thought that such sources account for a significant fraction of dioxin emitted. A recent EPA OAQPS report states that, due to the low chlorine level (0-100 ppm) in gasoline and diesel fuel, mobile sources may not be a significant source of dioxin.[76]

8.4 Current Activities

OAQPS plans to make a decision within the coming year on whether to list dioxin as a hazardous air pollutant under Section 112 of the Clean Air Act. Also, OAQPS has recommended that OMS consider a program to quantitatively measure dioxin emissions from mobile sources. OMS is presently considering whether work in this area would be useful and has asked for some additional OAQPS input on the relative priority of work on dioxins from mobile sources versus other sources.

9.0 ASBESTOS

9.1 Emissions and Ambient Concentrations

Asbestos, due to its high friction and heat resistance characteristics, is used in brake linings, clutch facings and automatic transmissions. About 22% of the total asbestos used in 1984 was used in motor vehicles. Previous EPA work involved measurement of asbestos emissions from vehicles during typical vehicle operations for a vehicle with front disc brakes and rear drum brakes.[77,78] This work showed that the asbestos emissions were 4 ug/mile and possibly as high as 28 ug/mile. It is estimated that the 4 ug/mile emission figure would result in a maximum annual average asbestos level in a central city area of about 0.25 ng/m^3.[79] This is in agreement with what would be predicted in a typical street canyon, using a method developed by Southwest Research Institute under EPA contract.[80] Urban asbestos levels from all sources in large cities show average readings of 2.6-5 ng/m^3 but average levels in New York range from 8 to 30 ng/m^3; localized asbestos levels in dense traffic can be somewhat higher. Thus, asbestos from mobile sources is responsible for 1-10% of the total asbestos although a higher fraction (7-70%) could result with the high emission factor.

9.2 Cancer Risk

The National Academy of Sciences (NAS) has estimated lifetime risks for persons in urban areas.[81] Based on the data in the NAS report, unit risk estimates range from 6.6 x 10^{-7} to 2.6 x 10^{-5} per ng/m^3 exposure (6.6 x 10^{-4} - 2.6 x 10^{-2} per ug/m^3). On an annual basis, this translates to 9 x 10^{-9} to 3.6 x 10^{-7} per ng/m^3 exposure (NAS assumed an average lifetime of 73 years).

Maximum annual average asbestos levels in urban areas due to motor vehicle emissions are estimated to range from 0.25 to 1.75 ng/m^3, based on estimated emission rates of 4 to 28 ug/mile. Using these exposure estimates in conjunction with the range of unit risks, and assuming an urban population of 180,000,000, the resulting cancer risk is estimated to range from 0.405-113.4 cancer incidences per year.

9.3 Current Activities

Due to the risk to the general population as well as the risk to workers exposed to asbestos, the EPA Office of Pesticides and Toxic Substances, in January 1986, has proposed regulations under Section 6 of TSCA to ban certain uses of asbestos (i.e., asbestos-cement pipes, flooring tiles, and asbestos clothing) and to allocate permits to mine and import asbestos which would restrict its remaining uses.[79] The

National Resources Defense Council petitioned EPA in 1984 to ban the use of asbestos in motor vehicles. However, EPA feels that effective substitutes are still not available for many applications of asbestos in brakes. EPA feels that the restrictions on use of asbestos will encourage the development of suitable replacements for brake linings. EPA is also considering a ban on asbestos friction products about 5 years after the final rules are promulgated. Presently, semi-metallic disc brake pads made without asbestos are used in about 85% of new domestic cars with front wheel drive. Also, some manufacturers are introducing aramid fiber instead of asbestos for disc brakes. Development of non-asbestos substitutes for drum brakes has not yet been as successful as for disc brakes due to concerns about durability and heat resistance. Still, one automobile manufacturer is using semi-metallic drum brake linings in its new minivans. Another has reported progress in developing a drum brake lining with aramid fiber.

The proposed regulations would involve EPA issuing permits for mining of asbestos in the U.S. and importation of asbestos and its products. The total amount of asbestos to be permitted under these permits for the first year after the regulation is promulgated would be 30% of the average amount of asbestos mined or imported yearly during the base period of 1981, 1982, and 1983. The amount of the asbestos used in the following years would decrease annually by 3% from the 30% level until it reached a 3% level in the tenth year of the regulation after which no asbestos could be mined or imported. Recent findings that there are not yet available good substitutes to replace asbestos in certain automobile and truck brakes, however, may push back EPA's goal for banning and phasing out asbestos for such uses.

10.0 VEHICLE INTERIOR EMISSIONS

10.1 Composition and Concentrations

Two different EPA projects were conducted measuring different compounds in vehicle interiors.[82,83]

One project showed vinyl chloride to be present at levels ranging from below 2 ppb to 7 ppb in several closed vehicles under high temperatures as might exist on a summer day.[82] Under similar conditions other carcinogenic compounds were identified qualitatively. The carcinogenic compounds detected are listed below.

aniline
biphenyl
1,2-dibromoethane
dichlorobenzene
dimethylphenol
isobutyl alcohol
maleic anhydride
naphthalene
benzene
carbon tetrachloride
chloroform
phenol

A total of 147 compounds were identified.

In another EPA project, 58 vehicle interiors were sampled for nitrosamine which is a potent carcinogen.[83] The main compound measured is N-nitrosodimethylamine which was found at levels of about 0.05 ug/m^3. This exposure level for 3 hours/day which would represent a long commute is less than that from a can of beer or a strip of bacon a day.

Also, from time to time, complaints have been received by EPA on the formation of a white semi-opaque film on the interior of a vehicle windshield. This film very slightly obstructs visibility and can be somewhat difficult though not impossible to remove. EPA ORD has analyzed samples of this film and finds it to consist of a phthlate ester; dioctyl phthlate is used as a plastisizer inside vehicles. The exposure level to and the health effects of this compound are not known.

10.2 Cancer Risk and Current Activities

At this point, no risk assessment calculations have been made for these vehicle interior emissions (with the exception of some simple calculations for the nitrosamines). Since the exposure level is low, it is thought that no significant risk could be occurring due to exposure to these substances. No further work is planned to identify other substances present in vehicle interiors at this time.

11.0 METALS

This section discusses metals which are emitted in vehicle exhaust. Lead, manganese , platinum and cadmium are considered. For each metal, available information regarding the source of the metal, emission rates and health effects are given. In addition, for cadmium, a risk estimate is presented.

11.1 Lead

11.1.1 Source and Emission Factors

Lead, in the form of tetraethyl lead and tetramethyl lead, is used in gasoline fuel to increase the octane number of gasoline and suppress knock. Knock is the premature autoignition and very rapid combustion of the fuel-air charge in the engine combustion chambers. Knock is frequently audible as a sharp metallic rap, and it may cause damage to engines. Lead alkyl compounds were first added to gasoline in 1923 as a means of suppressing engine knock by promoting uniform burning of the fuel-air mixture in the engine combustion chambers.

When lead alkyl compounds are burned, lead oxide is formed. To reduce the tendency of lead oxide to build deposits in automobile engines, halogen compounds are included in the fuel to scavenge the lead deposits from the engine. Ethylene dichloride and ethylene dibromide are commonly used. Analysis of lead particulate indicates that most of the exhausted lead appears as PbClBr.[71]

Vehicles equipped with catalytic converters are required to use unleaded gasoline since the presence of lead poisons the catalytic converter. Catalyst-equipped vehicles were introduced in 1975. Unleaded gasoline is subject to the requirement that it not contain any lead additives and that it not include more than 0.05 grams per gallon (gpg) lead.

Due to the widespread and persistent misuse of leaded gasoline in vehicles designed for unleaded gasoline (termed misfueling or fuel switching) and the adverse health effects of lead in gasoline, EPA promulgated regulations to reduce the lead content of gasoline.

In 1973, EPA required refiners to meet a 0.5 gpg standard for the average lead content of all gasoline. EPA later replaced this standard with a standard for the lead content of leaded gasoline only. Effective November 1, 1982, large refineries were required to meet a standard of 1.10 grams per leaded gallon (gplg). Certain smaller refineries were subject to a 1.90 gplg standard until July 1, 1983, at which time they would also be subject to the 1.10 gplg standard.[84] EPA further reduced the standard to 0.10 gplg effective on January 1, 1986, with an interim standard of 0.5 gplg effective on July 1, 1985.[85] EPA is now investigating the possibility of regulatory action banning the use of leaded gasoline. A range of alternatives is being considered, ranging from no ban to a ban in 1995 to a ban effective as early as January 1, 1988.[86]

Lead emissions from mobile sources are calculated based on the percentage of burned lead exhausted at different speeds, the lead content of gasoline, vehicle fuel economy and the model year mix of vehicles on the road. A report containing guidelines for predicting lead emission factors is available.[87] Based on the information in this report, simplified lead emission factors have been estimated for light-duty and heavy-duty model year 1986 (MY1986) vehicles. These emission factors are summarized in Table 11-1. Note that these estimates are for the mass of lead alone. To account for the halogens, these lead emission estimates should be multiplied by 1.557.[71]

Composite calendar year 1986 emission factors will vary from area to area depending on the model year mix of vehicles on the road, the driving conditions (which will affect fuel economy) and the misfueling rates in a particular area. Lead emission factors for heavy-duty gasoline-fueled vehicles are projected to decrease in later years since it is assumed that emission standards effective in 1987 will require virtually all new heavy-duty gasoline-powered vehicles under 14,001 pounds gross vehicle weight to use catalytic converters and thereby burn unleaded fuel.

11.1.2 Health Effects

A strong correlation has been demonstrated between gasoline lead usage and blood lead levels. The Centers for Disease Control (CDC) has defined 25 ug/dl as an elevated blood lead level. The list of demonstrated health effects at blood lead levels exceeding 30 ug/dl is well established.[88] Such effects include: 1) death at blood levels of 80+ ug/dl; 2) frank anemia, anorexia, abdominal pain, and vomiting at 70 ug/dl; 3) reduced hemoglobin at 40 ug/dl and interference in heme synthesis at levels down to 15-20 ug/dl; and 4) vitamin D metabolism interference at PbBs possibly as low as 12 ug/dl.

In addition, there is a great deal of evidence on neurological effects in children. At levels of 30+ ug/dl, studies have found significantly slowed nerve conduction, fine motor dysfunctions, impaired concept formation, lower IQ, and altered behavior among pre-school children. At PbBs as low as 15-30 ug/dl, a number of studies suggest possible small effects on IQ, behavioral dysfunctions (attentional deficits, poor classroom behavior), and changes in electrical brain wave activity and hearing function among children. Children appear

Table 11-1

Lead Emission Factors for MY1986 Vehicles

Vehicle	1986 Fuel Economy mpg*	1986 Lead Fuel Content gpg	Fraction of Lead Burned That is Exhausted	MY1986 Lead Emissions gpm
LDGV catalyst	23.8	0.014	0.75	0.0004
LDGV cat (misfueled)	23.8	0.10	0.44	0.002
HDGV	9	0.10	0.75	0.008
LDGV non-cat**	13	0.10	0.75	0.006

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* Combined city/highway fuel economy.

** This estimate is for a non-catalyst-equipped vehicle for comparison to the 1986 fleet. Fuel economy was based on data for pre-1975 light-duty gasoline-fueled vehicles.

to be particularly susceptible to adverse health effects from lead exposure. Recently emerging data from several ongoing longitudinal studies consistently indicate that fetal exposures to lead, at maternal or umbilical cord PbB levels as low as 10-15 ug/dl are associated with reduced birth weight and early growth, and delays in early mental, motor, and emotional development.

In addition, more recent data provide convincing evidence for strong associations between blood pressure increases and blood lead levels, even at blood lead levels below 30 ug/dl.

It is difficult to conclude what role lead may play in the induction of human tumors. Epidemiological studies of lead-exposed workers provide no definitive findings. Lead acetate has produced renal tumors in some experimental animals but does not seem to be a potent carcinogen.[89]

11.2 Manganese

11.2.1 Source and Emission Factors

A nonleaded organometallic additive which was used widely in the middle 1970's as an octane improver in unleaded gasoline is methycyclopentadienyl manganese tricarbonyl (MMT).

In September, 1978, EPA banned its use in unleaded gasoline because of evidence which showed that MMT increased hydrocarbon emissions and plugged catalysts.[90] This ban was temporarily suspended for four months (June through September) in 1979 because of concerns over potentially short supplies of unleaded gasoline.[91] MMT has always been allowed in leaded gasoline, but has been used at only low levels due to its high cost compared to lead additives.

More recently, a particulate trap regeneration system based on use of a manganese (Mn) containing fuel additive has been selected by Volkswagen as a means of meeting the 1986 California and 1987 Federal light-duty diesel particulate standard of 0.2 g/mile. EPA has given Volkswagen temporary approval to use this additive for the 1986-1988 model years.

Volkswagen estimates that for highway driving, emission rates will range from an expected value of 1.38 mg Mn/mile to a worst-case value of 4 mg Mn/mile. For street driving, expected emissions are 3.13 mg Mn/mile with a worst-case value of 10 mg Mn/mile.[92] The worst-case numbers are based on the assumption that all of the manganese is emitted in the exhaust while the expected numbers reflect expected manganese retention in the engine and particle trap. Data are presently lacking on the size distribution of particles emitted in the exhaust and the amount and species of manganese they contain.

The California Air Resources Board (CARB) estimates that, by 1990, between 2.6 percent and 6.6 percent of the California vehicle fleet will have traps. Using this estimate of trap usage, ambient Mn concentrations corresponding to highway emissions of 1.3 - 4 mg Mn/mile are estimated to be 0.03 - 0.24 ug Mn/m^3. Ambient concentrations corresponding to street canyon emissions of 3.13 - 10 mg Mn/mile are estimated to be 0.06 - 0.5 ug Mn/m^3. [92]

11.2.2 Health Effects

In June, 1984, Volkswagen requested that the Health Effects Institute (HEI) undertake an evaluation of potential health issues related to emissions from diesel automobiles using manganese-containing fuel additives for particle trap regeneration. This section briefly summarizes the highlights of HEI's October, 1985 response to Volkswagen.[92]

The effects of greatest concern are neurotoxic and respiratory. The neurotoxic effects in humans require at least several months of exposure and progress from an early reversible stage to a more advanced and irreversible stage. The human clinical and epidemiologic literature suggests that neurotoxicity is not strongly indicated until exposure exceeds 5 mg Mn/m^3, but that neurological symptoms may occur at levels as low as 0.3 mg Mn/m^3.

The respiratory effects of manganese are identical to those associated with exposure to fine or respirable particulate matter in general, and involve an inflammatory effect or pneumonitis, which may lead to diminished pulmonary function, bronchitis, or altered susceptibility to infection. EPA considers respiratory symptoms to be the critical effect because respiratory effects are reported at levels lower than those reported for neurotoxicity. EPA derived human No Observed Effect Levels (NOEL) of 5 ug Mn/m^3 based on rat data and 8.7 ug Mn/m^3 based on monkey data. [93]

Occupational studies generally show that respiratory effects follow exposures in excess of 5 mg Mn/m^3. However, data from a Japanese study of junior high school students exposed to manganese emissions from a ferromanganese plant apparently associates increased respiratory symptoms and diminished pulmonary function with exposures below 5 mg/m^3. Exposure was determined by the amount of manganese in the dustfall. EPA estimated, on the basis of analyses of dustfalls near a ferromanganese plant in the Kanawha Valley in West Virginia, that the dustfall in the Japanese study may translate to between 3 and 11 ug Mn/m^3. These are the lowest levels associated with adverse health effects; however, the correlation of settled dust with suspended particulate matter is subject to broad variability, even in the same geographic locations, as a result of air turbulence, humidity, topography, windspeed, etc. As a result, there is a great deal of uncertainty regarding the level of exposure to manganese concentrations associated with the health effects in this study. HEI is presently evaluating the study in detail.

There is some evidence of carcinogenic activity of manganese in laboratory animals, although the value of these studies is questionable. There is no epidemiologic information relating manganese exposure to cancer occurrence in humans.

11.3 Platinum

11.3.1 Source and Emission Factors

Platinum is a catalyst attrition product. It is emitted in small quantities. Most studies that have attempted to characterize platinum emissions report no detectable levels. The measurement methods used are often not sensitive enough to detect the small quantities of platinum emitted.

Limited quantitative measurements of platinum have been made. In 1978, Ford measured FTP particulate platinum emissions from two vehicles (1976 LTD with monolithic three-way catalyst, oxidation catalyst and air pump and a 1978 Pinto with monolithic three-way catalyst, oxidation catalyst and air pump). Platinum emissions ranged from 0.4 to 1.4 micrograms per mile (ug/mile).[94] In 1977, General Motors reported that platinum emissions from pelletized catalysts ranged from 1 to 3 ug/mile.[95]

It has been thought for some time that much of the platinum emitted may accumulate alongside roadways. This was confirmed in a recent study.[96] Dust samples collected from the leaves of roadside plants contained as high as 0.7 ppm of platinum and 0.3 ppm of palladium (another catalyst attrition product). The highest concentrations of both metals were found in dust collected from plants growing at the edge of heavily trafficked streets and highways and the lowest in samples collected from plants growing in the yards of houses located on lightly trafficked streets. The concentrations of both metals in the dust samples are much higher than the reported natural abundances of these elements.

11.3.2 Health Effects

In 1977, the National Academy of Sciences prepared a study which examined the uses, sources of supply, and potential health effects of noble metals. With regard to platinum and palladium emissions from catalyst-equipped vehicles, the NAS study concluded:

> "Minute quantities of platinum and palladium (about 1-3 ug/mile) are emitted from the exhaust systems of automobiles equipped with catalytic converters; much of this material may accumulate alongside roadways. However, this material is in a chemical form that is physiologically innocuous (no detectable soluble salts), and it is concluded that such emission poses no threat to the environment. Because there is no evidence that platinum metal can be methylated by microorganisms and solubilized in the same way that mercury is methylated, this deposited material should not have an adverse effect on the environment."[97]

Recently, Dr. Hans A. Nieper of the Paracelsus Clinic in West Germany has claimed that platinum emissions from catalyst-equipped vehicles are associated with rising incidences of AIDS, cancer and leukemia.[98] He also states that slight traces of platinum are extraordinarily toxic against the genetic defense systems. These claims are not currently scientifically supportable.

11.4 Cadmium

11.4.1 Source and Emission Factors

It is postulated that cadmium is present in fuel as a lead contaminant and therefore emitted by vehicles burning leaded fuel. Cadmium emissions from non-catalyst-equipped vehicles are roughly 1.6×10^{-5} g/mile.[32] Assuming 12 percent of the current light-duty fleet are non-catalyst-equipped vehicles, the resulting light-duty fleet emission factor is 1.9×10^{-6}.

11.4.2 Health Effects and Risk Estimate

A unit risk estimate for cadmium is estimated by EPA to be 1.8×10^{-3}. The risk from cadmium was determined by multiplying the risk determined in the Six Month Study by ratios of the updated to the Six Month Study emission factors and unit risks to obtain a crude estimate of the risk. The resulting risk of 0.001 per urban million appears negligible. This risk is further projected to decrease to zero by 1995 as non-catalyst-equipped vehicles are phased out of the fleet.

12.0 SIX MONTH STUDY: SUMMARY AND COMPARISON OF RESULTS

12.1 Purpose of Six Month Study and Summary of Results

The final study is formally titled, "The Air Toxics Problem in the United States: An Analysis of Cancer Risks for Selected Pollutants."[1] The EPA study attempted to assess the magnitude and nature of the air toxics problem by developing estimates of the cancer risks posed by selected air pollutants and their sources.

Three major analyses were used to estimate excess cancer risk from exposure to 15-45 toxic air pollutants. The Ambient Air Quality study used ambient data for five metals, 11 organic compounds, and benzo (a) pyrene (B(a)P) together with unit risks for these compounds to estimate national excess cancer incidence. The other two analyses (the NESHAPS study and the 35 County study) used exposure models together with unit risks to estimate excess cancer incidence. The NESHAPS study provides national estimates for about 40 compounds. The 35 County study was limited to 22 compounds and 35 counties but was designed to allow more detailed assessment of source contributions. Since the 35 County study was used to estimate the mobile source contribution to the air toxics risk, it will be discussed in more detail. The other two studies do not contradict any relevant findings from the 35 County study.

The 35 counties were chosen from counties with the highest expected ambient exposures, based on high populations and large aggregate emissions from all source categories. Also, some counties were included simply because they contain large industrial point sources of potential interest. The 35 counties contain roughly 20 percent of the total U.S. population (45 million people), 20 percent of total releases of VOC, and 10 percent of total particulate matter loading, based on 1982 data. They represent a variety of industrial and population distributions, but are not considered a statistically representative sample of the country.[99] When comparing the results of the 35 County study with the results of this study, cancer incidences will be expressed per million people exposed.

The aggregate cancer risk from all sources in the 35 County study is 207, or 4.6 cancer incidences per million, when products of incomplete combustion (PIC) are included. Mobile sources were estimated to account for 60 percent of the total incidence when PIC are included and 23 percent of the total incidence when PIC are excluded. A breakdown of the specific mobile source pollutants responsible and the risk associated with each pollutant is given in Table 12-1.

Table 12-1

Six-Month Study Results for Mobile Sources*

Pollutant	Total	Annual Cancer Incidence Per Million**	% Contribution
PIC	93.2	2.07	82.9
Benzene	14.0	0.31	12.5
Formaldehyde	3.1	0.07	2.7
Ethylene Dibromide	0.9	0.02	0.8
B(a)P	0.8	0.02	0.7
Cadmium	0.4	0.01	0.4
TOTAL:	112.4	2.50	100.0

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* Gasoline vapors were considered separately. The total risk was 6.8 or 0.15 per million.

** Based on 45 million people in 35 County study.

PIC refers to a large number of hydrocarbon compounds (mainly polynuclear organics). The PIC unit risk value is derived from various older epidemiological data and is controversial. Epidemiological studies of the general population exposed to ambient air and studies of workers occupationally exposed to PIC (e.g., coke oven emissions, hot pitch fumes) were used to estimate the PIC unit risk. B(a)P was used as an index of exposure to PIC. As a result, the PIC unit risk is expressed per unit of exposure of B(a)P. The PIC unit risk used in the Six Month Study is 4.2×10^{-1}. The uncertainties associated with this unit risk are detailed in the Six Month Study.

The B(a)P emission factor for mobile sources was used to calculate an annual average exposure. The B(a)P exposure was then multiplied by the PIC unit risk to estimate annual cancer incidence due to PIC from mobile sources. The same process was performed for all other sources emitting B(a)P to determine cumulative annual cancer incidence from PIC.

In the 35 County study, roughly 75 percent of the total B(a)P emissions was said to be attributable to mobile sources. Sources emitting the remaining 25 percent are residential fireplaces, woodstoves and waste oil burning, all considered area sources. Point sources were assumed to emit no B(a)P, since coke oven emissions were dealt with separately in the report. As a result, 75 percent of the total PIC cancer incidence is due to mobile sources. PIC cancer incidence accounts for a large portion of the aggregate cancer incidence because the unit risk for PIC is orders of magnitude greater than the unit risks for the other toxic compounds examined in this study. This explains why, when PIC are included, the fraction of total cancer incidence related to mobile sources increases from 23 to 60 percent.

From Table 12-1, the pollutants responsible for the total estimated cancer incidence from motor vehicles in the 35 County study, in order of importance, are PIC, benzene, formaldehyde, ethylene dibromide (EDB), B(a)P and cadmium. Gasoline vapors from motor vehicles were not included in the motor vehicle category. Instead, gasoline vapors from service stations, which includes vehicle refueling and delivery of gasoline, were considered separately.

12.2 Comparison of Six Month Study Results with Results of This Study

For benzene, formaldehyde, gasoline vapors and 1,3-butadiene, the annual cancer incidence per million estimated in both studies can be directly compared. For PIC, the PIC risk estimated in the Six Month Study will be compared to the combined risk from diesel particulate and gasoline POM/PIC estimated in this study. For EDB and cadmium, risks have not been estimated in this report due to the rather low emissions and the lack of data on emissions at various speeds required as inputs to the NEM model. For these pollutants, rough estimates of the risk will be made by comparing the emission factors used in the Six Month Study with those which will be developed in the subsequent sections, and adjusting the risks obtained in the Six Month Study accordingly. This approach assumes that the emissions are directly associated with ambient concentrations. This appears to have some merit, considering that EDB and cadmium are not considered photochemically reactive.

In addition to emission factors, the unit risks used in both studies should be compared. In some cases, the unit risks have changed substantially since release of the Six Month Study. 1,3-Butadiene is one notable example.

In the following sections, the risks from each pollutant will be compared. The emission factors and unit risks used to calculate the annual cancer incidences will also be compared in order to pinpoint the discrepancies. Finally, the aggregate risks will be compared. Table 12-2 presents a summary of this comparison that will be referred to in the following subsections.

12.2.1 Formaldehyde

Before comparing the composite emission factors for formaldehyde, the vehicle class VMT fractions used will be briefly discussed. For this study, the VMT fractions are those used in MOBILE3 for calendar year 1986. For the Six Month Study, the supporting document prepared by Versar for the 35 County study included VMT data for only one of the 35 counties.[99] Also, the emissions and VMT data in the 35 County study were grouped into three major vehicle categories (LDG, HDG and HDD). The MOBILE3 VMT fractions were grouped into these three vehicle categories and compared to the VMT fractions for the single county. The VMT fractions were generally similar; the MOBILE3 heavy-duty VMT fractions were somewhat higher. As a result, the MOBILE3 grouped VMT fractions were applied to the emissions data for LDG, HDG and HDD given in the Versar report to calculate composite emission factors for the 35 County study.

As seen in Table 12-2, both the composite emission factor and the unit risk for formaldehyde used in this report are higher than those used in the 35 County study (referred to as the Six Month Study in Table 12-2). The emission factors used in the 35 County study were those derived from studies of low mileage vehicles. In this study, the emission factors were percentages of total exhaust HC, as predicted by MOBILE3 for 1986, and were designed to account for in-use deterioration. Also, since the release of the Six Month Study, the unit risk

Table 12-2

Comparison of Results

Pollutant	Composite Emission Factor (g/mile) 6 Month	Composite Emission Factor (g/mile) This Study	Unit Risk 6 Month	Unit Risk This Study	Annual Mobile Source Cancer Risk (per urban million) 6 Month	Annual Mobile Source Cancer Risk (per urban million) This Study[a]
Formaldehyde	0.033	0.042-0.045	6.1×10^{-6}	1.3×10^{-5}	0.07	0.22-0.70
Benzene	0.131	0.100	6.9×10^{-6}	8.0×10^{-6}	0.31	0.44-1.0
PIC[b]	1.66×10^{-5}	ND[c]	4.2×10^{-1}	ND	2.07	ND
B(a)P	1.66×10^{-5}	d	3.3×10^{-3}	3.3×10^{-3}	0.02	d
Diesel Particulate	ND	d	ND	$0.20\text{-}1.0\times10^{-4}$	ND	0.76-3.67
Gasoline PIC/POM	ND	d	ND	d	ND	0.007-0.76
Gasoline Vapors			7.5×10^{-7}	1.18×10^{-6}	0.15	0.36
1,3-Butadiene	1.0×10^{-3}	0.024-0.026	4.6×10^{-7}	2.8×10^{-4}	0	0-3.16
Ethylene	ND	0.237-0.261	ND	2.7×10^{-6}	ND	0-0.29
Asbestos	ND	$4\text{-}28\times10^{-6}$	ND	$6.6\text{-}260\times10^{-4}$	ND	0.002-0.63
Cadmium	8.5×10^{-6}	1.9×10^{-6}	2.3×10^{-3}	1.8×10^{-3}	0.01	0.001
Ethylene Dibromide	1.3×10^{-4}	7.1×10^{-5}	5.1×10^{-4}	5.1×10^{-4}	0.02	0.01
				TOTAL:	2.65	1.80-10.58

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[a] An urban population of 180 million was used.

[b] Diesel particulate and particle-associated organics from gasoline-fueled vehicles were considered to represent PIC in this study. Therefore, a PIC risk was not determined in this study.

[c] ND=Not Determined.

[d] See text for explanation.

for formaldehyde has increased substantially. The unit risk used in this report is an upper 95 percent confidence limit and includes only malignant tumors.

The annual cancer risk for both studies assume an average lifetime of 70 years. The risk per million for the 35 County study is based on a population of 45 million. This population was assumed to be urban. The calendar year 1986 annual cancer risks in this study presented in Table 12-2 are based on an estimated 1986 urban population of 180 million.

When the annual cancer risk for formaldehyde given in the 35 County study is adjusted to account for the updated unit risk estimate, the resulting annual cancer risk is 0.15 per million. This still falls below the low end of the range given in this study. The range (0.22-0.70 per urban million) attempts to account for formaldehyde photochemistry. The range is large because 1) it is difficult to determine what the annual average formaldehyde concentration is for any particular area, much less the entire urban population, and 2) the relative contribution of mobile and stationary sources is not known. For example, the low estimate (0.22 per urban million) is based on a NEM-predicted concentration of 1.21 ug/m^3 for direct emissions of formaldehyde from mobile sources. The highest estimate (0.70 per urban million) assumes a maximum annual average concentration of 12.7 ug/m^3 based on monitoring data, with 30 percent attributable to mobile sources, and attempts to account for formaldehyde photochemistry.

From comparing the two risk estimates (the 35 County estimate and the low estimate in this study) and accounting for differences in the emission factors and unit risks, it appears that the NEM used in this study predicts slightly higher exposures per g/mile emitted than the dispersion modeling used in the 35 County study. This should be kept in mind when comparing the results for benzene, diesel particulate, gasoline POM/PIC and 1,3-butadiene as well since the modified NEM was used to estimate exposures for these pollutants.

12.2.2 Benzene

As seen in Table 12-2, the risks for benzene are more similar; however, the risk obtained in the Six Month Study still falls below the range of risk estimates determined in this study. The low estimate in this study is based on a NEM-predicted concentration whereas the high estimate is based on available ambient monitoring data, assuming 85 percent is attributable to mobile sources. The 35 County study used greater emission factors but a lower unit risk. Like formaldehyde, the unit risk for benzene has increased since release of the Six Month Study.

12.2.3 PIC, B(a)P, Diesel Particulate and Gasoline PIC/POM

The Six Month Study calculated a risk estimate for products of incomplete combustion (PIC). The approach used has been previously described. Benzo (a) pyrene (B(a)P) was treated separately. In this study, information existed to enable separate treatment of diesel and gasoline PIC. Diesel particulate was considered to represent diesel PIC while particle-associated organic emissions from gasoline-fueled vehicles were considered to represent gasoline PIC. The sum of these risks will be compared to the PIC risk obtained in the Six Month Study.

The risk from diesel particulate ranges from 0.76 - 3.67 per urban million. The range is due to the range of unit risks chosen.

For gasoline-fueled vehicles, three different approaches were taken to estimate the risk from gasoline PIC. These approaches are discussed in detail in Section 7. The first approach estimates the risk of B(a)P from gasoline-fueled vehicles. B(a)P emission factors are used together with the B(a)P unit risk. The annual cancer risk of B(a)P from gasoline-fueled vehicles was determined by multiplying the B(a)P risk obtained in the Six Month Study by the ratio of the emission factors (this study / Six Month Study). This approach assumes that there is no risk from the remaining gasoline PIC emissions.

The composite B(a)P emission factor of 5.56×10^{-6} g/mile used in this study is roughly one-third of the emission factor used in the Six Month Study (1.66×10^{-5}). The main reason for this difference is that the Six Month Study assumed 50 percent of the gasoline-fueled vehicles were leaded while this study used a more realistic percentage of 12 percent.

When the B(a)P risk estimated in the Six Month Study (0.02 cancer incidences per urban million) is multiplied by the ratio of the composite emission factors ($5.56 \times 10^{-6}/1.66 \times 10^{-5}$), the resulting annual cancer risk is 0.007 per urban million.

The second approach uses B(a)P emission factors from gasoline-fueled vehicles together with the PIC unit risk (which is expressed per unit of exposure of B(a)P) to estimate the annual cancer risk of PIC from gasoline-fueled vehicles. This is the approach used in the Six Month Study. The annual cancer risk of PIC from gasoline-fueled vehicles was determined by multiplying the PIC risk obtained in the Six Month Study by the ratio of the B(a)P emission factors.

When the PIC risk estimated in the Six Month Study (2.07 per urban million) is multiplied by the ratio of the composite emission factors ($5.56 \times 10^{-6}/1.66 \times 10^{-5}$), the resulting annual cancer risk is 0.68 per urban million.

Unlike the second approach, the third approach does not use B(a)P as a surrogate for PIC emissions. The third approach uses estimated emission rates of gasoline particle-associated organics (as an unspeciated mixture) together with a unit risk for these mixed organics. Exposures were estimated using the modified NEM.

Estimated composite emission factors for gasoline particle-associated organics in 1986 are estimated to be 0.0075 - 0.0082 g/mile, depending on whether a minimum I/M program was assumed to exist. An upper confidence limit unit risk is 2.5×10^{-4}.

Nationwide urban exposure in 1986, using the modified NEM is estimated to be 0.20-0.21 ug/m^3. Estimates of urban lung cancer incidence in 1986 range from 127-136, or 0.71-0.76 per urban million.

The resulting range of risk estimates for gasoline PIC, using the results of all three approaches, is 0.007-0.76 per urban million.

The sum of diesel particulate and gasoline POM/PIC risks, or equivalent "PIC" risks, range from 0.77-4.43 per urban million. The PIC risk obtained in the 35 County study (2.07 per urban million) lies within this range.

12.2.4 Gasoline Vapors

The annual cancer risk from gasoline vapors reported (based on other documents) in this study is 0.36 per urban million. In the Six Month Study, the risk from gasoline marketing was considered. Gasoline marketing in the Six Month Study refers to emissions from service stations. It includes emissions from vehicle refueling and from the delivery of gasoline to the service station. The resulting risk was 0.15 per million. This is less than half the risk presented in this study. It is mentioned, however, in the supporting Versar report that high exposures near the pump from self-service refueling were excluded. The vehicle refueling in the Six Month Study then refers to occupational exposure (service station attendants). The risk presented in the Six Month Study is, as a result, more in line with the risk presented in this study, since the risk from self-service refueling was projected to be 54 percent of the total.

12.2.5 1,3-Butadiene

The 35 County study found no risk associated with emissions of 1,3-butadiene from mobile sources. A cancer risk estimate for 1,3-butadiene was estimated in this study using an updated emission factor and unit risk. The risk estimate is discussed in detail in Section 6.

Determination of an emission factor for 1,3-butadiene is difficult. This is because 1,3-butadiene and n-butane coelute and thus have the same retention point on the gas chromatograph. Emission characterization studies to date have not attempted to determine the percentage of the peak due to 1,3-butadiene. Therefore, assumptions must be made about the percentage each compound contributes to this peak. It will be assumed that 15 percent of the peak is due to 1,3-butadiene.[64] 1,3-Butadiene emissions were expressed as a percentage of the total exhaust HC predicted by MOBILE3. Based on the data from 46 in-use gasoline-fueled vehicles provided in reference 30, 1,3-butadiene is roughly 0.94 percent of the total FID exhaust HC. Due to the lack of data for the other vehicle classes, this percentage was simply applied to the MOBILE3 composite exhaust HC emission factor.

The modified NEM model was used to estimate exposures. Nationwide urban exposure in 1986 is estimated to be 0.69-0.76 ug/m^3. The range accounts for both the presence and absence of an Inspection/Maintenance program. These exposure estimates are for direct emissions of 1,3-butadiene and do not account for reactions of 1,3-butadiene in the atmosphere. Available ambient monitoring data were reviewed and compared to the exposure estimates. Average mean values in urban settings range from 0.77-24.23 ug/m^3.[64] The NEM estimate of urban exposure from motor vehicles lies near the low end of this range.

The unit risk for 1,3-butadiene has changed substantially. Based on inhalation studies of 1,3-butadiene in mice, a 95 percent upper confidence limit unit risk for 1,3-butadiene is estimated to be 2.8×10^{-4}.[66] This is much greater than the previous estimated unit risk of 4.6×10^{-7} used in the Six Month Study.

Estimates of urban cancer incidence in 1986 range from 514-568, or 2.86-3.16 per urban million. Due to the uncertainty associated with the emissions estimate, a lower risk estimate of zero will also be used.

12.2.6 Ethylene

Ethylene from motor vehicles was not considered in the 35 County study. Based on the unit risk for ethylene provided in the Six Month Study, risk estimates for ethylene were computed in this study (Section 6.4). The urban risk in 1986 was estimated to be 47-52 cancer incidences, or 0.26-0.29 per urban million. As mentioned in Section 6.4, however, the unit risk and resulting risk estimates must be regarded as extremely tentative, since there is no available evidence that ethylene is carcinogenic. The unit risk for ethylene was estimated based on assumptions regarding its potency relative to ethylene oxide, a metabolite of ethylene and an animal carcinogen. As a result, a lower risk estimate of zero is also used.

12.2.7 Asbestos

Asbestos emissions from motor vehicles were not considered in the Six Month Study. Based on available emission factors and resulting expected ambient concentrations, it is estimated in this study that asbestos emissions from motor vehicles could currently be responsible for as many as 0.002-0.63 cancer incidences per urban million.

12.2.8 Ethylene Dibromide (EDB) and Cadmium

EDB and cadmium were handled similarly. Updated emission factors and unit risks were used in this study. For each pollutant, the risk determined in the 35 County study was multiplied by ratios of the updated to the 35 County emission factors and unit risks to obtain a crude estimate of the risk.

EDB is used as a lead scavenger. It is therefore emitted from vehicles using leaded gasoline. EDB data came from reference 100. The light-duty vehicle fleet was assumed to be 88 percent catalyst-equipped and 12 percent non-catalyst-equipped based on MOBILE3 data. Of the catalyst-equipped vehicles, a misfueling rate of 14.5 percent was used.[87] The composite emission factor for light-duty vehicles is 7.1×10^{-5} g/mile. Heavy-duty gasoline-fueled vehicles were not considered. The impact of this vehicle class is not expected to be great due to its small contribution to total VMT (4 percent).

The composite emission factor and resulting annual cancer risk is roughly half that calculated in the 35 County study. This is because a much higher percentage of non-catalyst-equipped vehicles was assumed in the 35 County study (50 versus 12 percent). The resulting risk of 0.01 per urban million appears negligible.

In 1995, with the phase-out of leaded-fueled vehicles, the emission factor is expected to decrease to 1.8×10^{-5} g/mile. The resulting risk is 0.003 per urban million.

Data for cadmium were obtained from references 32-35. Cadmium does not appear to be emitted from catalyst-equipped vehicles or it is emitted below the detection limit. Cadmium emissions from non-catalyst-equipped vehicles are roughly

1.6×10^{-5} g/mile. It appears that cadmium may be a lead contaminant. Assuming 12 percent of the light-duty fleet are non-catalyst-equipped vehicles, the resulting light-duty fleet emission factor is 1.9×10^{-6}. Again, this is less than what was used in the 35 County study because of the lower percentage of non-catalyst-equipped vehicles expected. The resulting risk of 0.001 per million is negligible compared to the other risks.

12.2.9 Total Aggregate Risk

The sum of all the individual annual cancer risk estimates in the 35 County study (including gasoline vapors) is 2.65 per million. In this study, the aggregate risk ranges from 1.80 to 10.58 per urban million. If the formaldehyde risk in the 35 County study is increased to reflect the updated unit risk, the aggregate risk in the 35 County study would increase to 2.73 per million. The range in this study is due to a number of factors including: 1) the uncertainty of annual average formaldehyde and benzene concentrations and the contribution of mobile sources, 2) the range of unit risks used for diesel particulate, and 3) the different approaches used to determine the contribution of gasoline POM/PIC.

It should be noted when reviewing Table 12-2 that the pollutants given do not represent a complete list. This list does not include pollutants which are formed photochemically from mobile source emissions. This category of pollutants could have considerable impact but not enough is known to make a quantitative estimate.

13.0 SUMMARY AND LIMITATIONS

The aggregate risk from mobile source pollutants in calendar year 1986 was estimated to range from 1.80-10.58 per urban million. This translates into roughly 325-1900 urban cancer incidences. The majority of risk is attributed to formaldehyde, diesel particulate, and benzene. Due to increasing use of advanced control technology, the risk in 1995 is projected to decrease to roughly 60 percent the risk in 1986.

When reviewing these estimates, the following important limitations should be considered.

- This report only accounts for a small number of mobile source pollutants known to be emitted. In reality, mobile sources emit hundreds of compounds. A combination of health and/or exposure and/or emissions data are lacking for many of these compounds.

- With the exception of formaldehyde, this report does not consider reactions of mobile source pollutants in the atmosphere. Little data are available. Resulting pollutants may be more or less carcinogenic than what was originally emitted.

- Exposures for many of the pollutants were estimated using a modified version of the NAAQS Exposure Model (NEM) for CO. In order to apply this model to other mobile source pollutants, it was assumed that the other pollutants have the same dispersion characteristics as CO. This is probably not true for every pollutant; however, this was determined to be the best approach currently available to determine exposure to mobile source pollutants on a national scale.

  An alternative is to use ambient data. Unfortunately, ambient data for many mobile source pollutants, particularly VOC, are scarce and not of high quality. In addition, the relative contributions of mobile and stationary sources are not known.

- This report does not fully take into account seasonal variations in emissions. Hydrocarbon emissions from mobile sources are known to increase as the temperature decreases. Work is being done to characterize emissions of formaldehyde under cold temperature conditions. This will be followed by further characterization of individual HC emissions from mobile sources under cold temperature conditions.

- The unit risk estimates are always subject to some uncertainty. With the exception of benzene, they are generally based on the results of animal rather than human studies.

- The risks are assumed to be additive. It may be that certain combinations of exposures have synergistic or antagonistic effects.

- The risk projections for 1995 are based on the emission standards currently in place. Changes in fuel composition are not considered.

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GLOSSARY OF TERMS

Airshed: A geographical area which, because of topography, meteorology, and climate, shares the same air mass.

Air Toxic: A compound in the air capable of causing adverse health effects. For the purpose of this report, the air toxics examined were limited to known or suspected carcinogens.

Aldehydes: A class of fast-reacting organic compounds containing oxygen, hydrogen, and carbon. They contain the group -CHO.

Aliphatic: A class of hydrocarbon compounds which are open chained and fully saturated (e.g., no double bonds).

Ambient Air: That portion of the atmosphere, external to buildings, to which the general public has access.

Ames Test: A mutagenicity bioassay using the bacteria, Salmonella typhimurium.

Aromatic: A class of hydrocarbon compounds originating from benzene, C_6H_6, or containing at least one benzene ring or similar unsaturated heterocyclic ring.

Benzo(a)Pyrene (B(a)P): A polycyclic aromatic hydrocarbon with the molecular formula $C_{20}H_{12}$.

Bioassay: Using living organisms to measure the effect of a substance, factor, or condition.

Catalyst: Used in this report to denote a catalytic converter, a chamber in the exhaust system of vehicles containing a catalyst system which aids in oxidizing the carbon monoxide and unburned hydrocarbons in the exhaust gases or in reducing nitrogen oxides in the exhaust gases to innocuous products (carbon dioxide, N_2, O_2, and water).

Control Technology: A combination of measures designated to achieve the aggregate reduction of emissions.

Diesel Particulate Trap: A device located in the exhaust stream of a diesel vehicle that filters a certain percentage of exhaust particulate. This device must include some means by which accumulated particulate can be burned, thus regenerating the trap and making the trap available for continued particulate filtration.

Dynamometer: A device that is used to measure or to simulate loads, engine torque, and driving forces on vehicles or engines.

Exhaust Gas Recirculation (EGR): A system or device (such as modification of the engine's carburetor or positive crankcase ventilation system) that results in engine operation at an increased air-fuel ratio so as to achieve reductions in exhaust emissions of nitrogen oxides.

Emission Factor: For motor vehicles, an emission factor is the amount of pollutant emitted per unit of distance. In this report, emission factors are expressed in units of grams of pollutant per mile travelled.

Epidemiology: The study of diseases as they affect populations rather than individuals, including the distribution and incidence of a disease; mortality and morbidity rates; and the relationship of climate, age, sex, race and other factors.

Evaporative Emissions: Hydrocarbons emitted into the atmosphere from a motor vehicle through fuel evaporation.

Exhaust Emissions: Substances emitted to the atmosphere from any opening downstream from the exhaust port of a motor vehicle engine.

Federal Test Procedure (FTP): A multistage (multimodal) test procedure for new car certification by the Environmental Protection Agency.

Halogen: Any one of the nonmetallic elements chlorine, iodine, bromine, and fluorine.

Heavy-Duty Vehicle: Any motor vehicle rated at more than 8,500 pounds gross weight or that has a vehicle curb weight of more than 6,000 pounds or that has a basic vehicle frontal area in excess of 45 square feet.

Hydrocarbon: Any of a vast family of compounds containing carbon and hydrogen in various combinations: found especially in fossil fuels.

Ketone: An organic compound derived by oxidation from a secondary alcohol; it contains the carbonyl group (= CO).

Light-Duty Vehicle: A passenger car or passenger car derivative capable of seating 12 passengers or less.

MOBILE3: A computer program that calculates emission factors for hydrocarbons (HC), carbon monoxide (CO), and oxides of nitrogen (NOx) from highway motor vehicles.

Mutagenic: The property of a substance or mixture of substances to induce changes in the genetic structure in subsequent generations.

NAAQS Exposure Model (NEM): An exposure model suitable for evaluating alternative ambient air standards.

Neurotoxic: Harmful to nerve tissue.

Noble Metal: Metal such as platinum or palladium that is non-reactive to most chemical substances.

Organic: In chemistry, any compound containing carbon.

Oxidation Catalyst: A catalytic converter used to oxidize the carbon monoxide and unburned hydrocarbons in the exhaust gases to innocuous products.

PAH: Polynuclear aromatic hydrocarbons.

Particulate: A particle of solid or liquid matter.

Photochemistry: Chemical changes brought about by the radiant energy of the sun acting upon various polluting substances. The products are known as photochemical smog.

POM: Polycyclic organic matter. Many POM compounds are also PAH compounds.

PIC: Products of Incomplete Combustion.

Soluble Organic Fraction (SOF): In this report, defined as the organic fraction of particulate soluble with dichloromethane.

Steady-State: Constant operating conditions with no variation in fuel supply or load.

Street Canyon: A street lined with buildings. In general, the streets are less than seven lanes wide and the buildings under 26 stories. The minimum canyon height to width ratio for a building-lined street to be considered a street canyon is approximately 0.3.

Three-Way Catalyst: A catalytic converter that is capable of both oxidizing carbon monoxide and unburned hydrocarbons and reducing nitrogen oxides in the exhaust.

TSCA: Toxic Substances Control Act.

Unit Risk: The individual life time excess cancer risk from continuous exposure to 1 ug carcinogen per m^3 inhaled air.

VMT: Vehicle miles travelled.

Volatile Organic Compounds (VOC): Any compound containing carbon and hydrogen or containing carbon and hydrogen in combination with any other element which has a vapor pressure of 1.5 pounds per square inch absolute or greater under actual storage conditions.